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Spectroscopy of excited states in $In_{0.53}Ga_{0.47}As$-InP single quantum wells grown by chemical-beam epitaxy

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Photoluminescence excitation (PLE) spectra are reported for six single quantum wells with thicknesses between 130 and 23 Å grown on the same wafer by chemical-beam epitaxy. The very strong, narrow line emission from these high-quality quantum wells enabled us to perform PLE with a lamp-monochromator combination as the excitation source. All of the observed excitonic absorption peaks are assigned. Good fits to the spectra can be made with band offsets of $Q_e \approx 60\%$ and $Q_h \approx 40\%$ and masses $m_e^* = 0.041m_0$, $m_{hh}^* = 0.465m_0$, and $m_{lh}^* = 0.085m_0$. Energy-dependent corrections for m_e^* due to conduction-band nonparabolicities are essential for these fits and yield $\gamma_e = 3.3\times10^{-15}$ cm^2 for the $\gamma_e k^4$ correction term in the energy dispersion.

Quantum wells (QW's) made from $In_{0.53}Ga_{0.47}As$ lattice matched to InP have become interesting semiconductor structures in the past few years both for basic research and for optoelectronic devices. The wavelength of the light emitted by such structures can be varied between ~1.55 μm (corresponding to the band gap of bulk $In_{0.53}Ga_{0.47}As$, ~0.8 eV) and 1.1 μm by variation of the thickness L_z of the wells.[1-3] Hence, they are ideal for optical communication systems using silica fibers. $In_{0.53}Ga_{0.47}As$ QW's have been prepared by many growth techniques including metal-organic chemical-vapor deposition (MOCVD),[4,5] chloride transport vapor-phase epitaxy (VPE),[6,7] and molecular-beam epitaxy (MBE) using solid arsenic and phosphorus sources[3,8,9] and arsine and phosphine sources.[10]

Recently, a new epitaxial technique has been developed, chemical-beam epitaxy (CBE).[11] It was shown[12] that $In_{0.53}Ga_{0.47}As$ QW's prepared by this method are of extremely high quality, superior in many respects to those made by MOCVD, VPE, or MBE. The optical emission from these QW's is intense and of narrow linewidth.[13]

Compared to $Al_xGa_{1-x}As$-GaAs quantum wells, relatively few studies have been reported on $In_{0.53}Ga_{0.47}As$ quantum wells. In particular, no photoluminescence excitation spectroscopy (PLE) has been reported on $In_{0.53}Ga_{0.47}As$ quantum wells, although such data could give valuable information on important parameters such as band offsets and relevant electron and hole masses. The lack of PLE data is partly due to the limited availability of tunable lasers over the wide spectral range from 1.1 to 1.6 μm. For QW's of $In_{0.53}Ga_{0.47}As$-$In_{0.52}Al_{0.48}As$ lattice matched to InP with widths down to 100 Å, PLE measurements have been reported in the spectral range from 1.42 to 1.59 μm using a tunable KCl:Tl color-center laser.[14] A PLE spectrum of a solid-source MBE-grown $In_{0.53}Ga_{0.47}As$ quantum well with $L_z \approx 80$ Å observed with lamp-monochromator excitation was very recently reported but no assignments of the many structures in the spectrum were advanced.[15] PLE experiments with a lamp-monochromator excitation source performed on a multi-quantum-well structure prepared by chloride VPE did not give clear evidence of excited states, and only the onset of absorption close to the position of the $n=1$, electron-to-heavy-hole transition was observed.[16]

In the present Rapid Communication we take advantage of the narrow and extremely intense emission of excitons confined to $In_{0.53}Ga_{0.47}As$ quantum wells grown by the CBE technique. Our PLE spectra were excited by a 100-W halogen lamp in conjunction with a 0.3-m McPherson grating monochromator at a blaze wavelength of 1.6 μm. The photoluminescence (PL) light was detected at 90° by a 0.6-m Jobin-Yvon grating single monochromator with 1.5 μm blaze wavelength. Since no double monochromators were employed care was taken to avoid artifacts in the spectra due to scattered light on the excitation or detection side. The PL light was detected by a cooled Ge diode and the signals processed by conventional lock-in techniques.

We focus here on PLE spectra from a seven-layer quantum-well structure grown on an InP(Fe) substrate wafer. Next to the substrate a 0.5-μm InP buffer layer was grown followed by a 1520-Å-wide $In_{0.53}Ga_{0.47}As$ control layer, and six wells of thickness $L_z = 130$, 80, 60, 40, 25, and 17 Å, as determined by transmission-electron-microscopy (TEM) cross-sectional micrographs. The InP barriers separating the wells are 600 Å thick. The control layer serves as a reference for the PL spectra from which precise energy upshifts of the QW emission can be calculated.

Figure 1 shows a 2-K PL spectrum of this sample at a few microwatts of 514.5-nm Ar laser line excitation. The line positions are, in consecutive order, 1.5116 μm (0.8200 eV) ($L_z = 130$ Å), 1.438 μm (0.8619 eV) (80 Å), 1.4114 μm (0.8782 eV) (60 Å), 1.3632 μm (0.9092 eV) (40 Å), 1.2634 μm (0.9811 eV) (25 Å), and 1.1974 μm (1.0351 eV) (17 Å). The linewidths are, in the same order, 3.6, 4.0, 5.2, 7.2, 10.6, and 11.8 meV. The emission of the control layer is at 1.5828 μm (0.7831 eV), virtually identical with bulk $In_{0.53}Ga_{0.47}As$.

Figure 2 shows PLE spectra obtained by PL light detec-

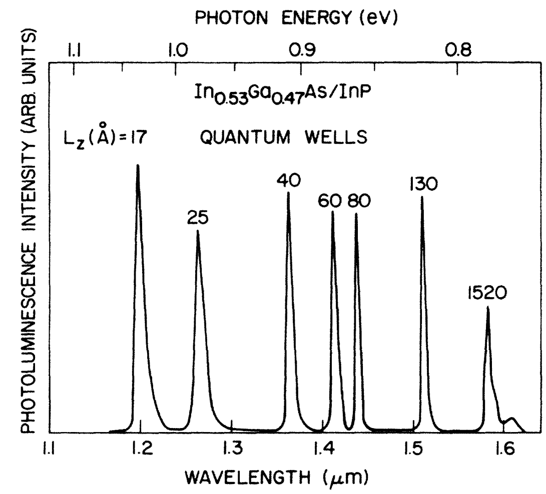

FIG. 1. Photoluminescence spectrum of the seven-layer sample at 2 K, excitation by a few μW of the 514.5-nm Ar laser line. Excited was a large area, $\sim 1.5\times1.5$ mm^2. Well thicknesses L_z refer to TEM analysis.

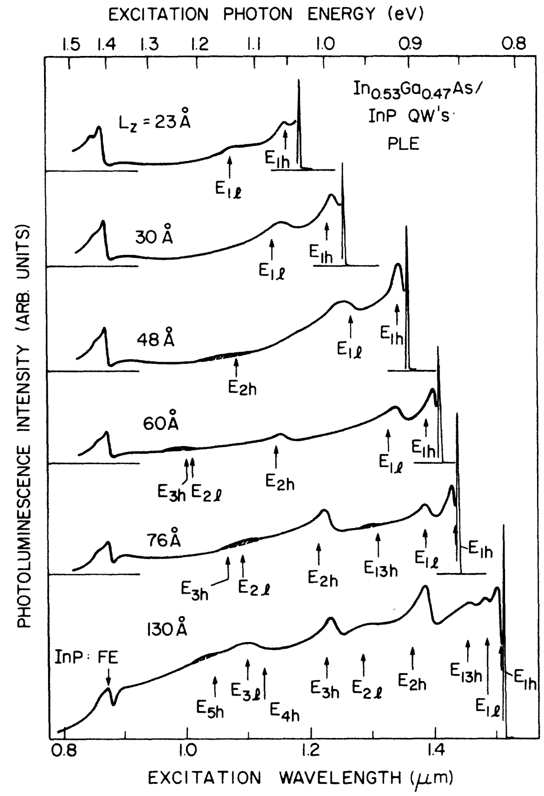

FIG. 2. Photoluminescence excitation (PLE) spectra of the multilayer sample at 2 K. Light detection is at the PL energy positions. An area of $\sim 1.5\times1.5$ mm ^{2} is excited. Weak peaks are shown hatched for clarity. The arrows and peak assignments refer to the best fit with $m_e^*=0.041m_0$, $\gamma_e=3.3\times10^{-15}$ cm^2, $m_{hh}^*=0.465m_0$, $m_{lh}^*=0.085m_0$, and $Q_e=60\%$ ($Q_h=40\%$). L_z values given are from these fits.

tion at these positions while scanning the excitation light to higher energies. The sharp peaks at the low-energy cutoffs of the spectra result when the exciting light scans over the fixed detection energies. The attenuation factors not shown on Fig. 2 range from 300 to 1400. All original recorder traces are almost free of noise. The resolution of the spectra is 1.2 nm, about half the widths of the sharp peaks, given by the convolution of the excitation and detection spectral functions. The bandwidths were chosen almost equal for both monochromators. At the high-energy cutoff of the spectra the free-exciton (FE) absorption peak of InP is observed at 0.8740 μm (1.4181 eV). The decrease of the signal toward higher energies above the InP band edge is ascribed to the sharp drop of the penetration depth of the excitation light into the sample. Compared to the best GaAs-$Al_xGa_{1-x}As$ QW's large Stokes shifts between exciton emission and absorption are evident from Fig. 2. The shifts are 6.0 meV ($L_z=130$ Å), 6.0 meV (76 Å), 6.6 meV (60 Å), 10.1 meV (48 Å), 14.3 meV (30 Å), and 24.3 meV (23 Å). Values of that order seem to be consistent with shifts observed between PL and photoconductivity (PC) spectra of MOCVD $In_{0.53}Ga_{0.47}As$ QW's of 75 and 110 Å well width (Fig. 2 of Ref. 17). PLE exciton absorption spectra as in Fig. 2 have been observed in our study from many other CBE-grown samples including multiple-quantum-well and single-quantum-well structures.

An assignment of our confined exciton peaks in Fig. 2 is suggested by a comparison to GaAs-$Al_xGa_{1-x}As$ ($x=0.5$) quantum wells grown by CBE.[18] In the latter system, for L_z in the range from 100 to 150 Å, exciton transitions up to E_{3h} ($n=3$ electron$\rightarrow n=3$ heavy hole) were observed with intensities much larger for the heavy-hole transitions than for the light-hole transitions, and relatively strong "forbidden" transitions E_{13h} ($n=1$ electron$\rightarrow n=3$ heavy hole), but no E_{12} transitions either for heavy or light holes. In view of similar electron and hole masses and comparable band-gap discontinuities of the GaAs-$Al_{0.5}Ga_{0.5}As$ and $In_{0.53}Ga_{0.47}As$-InP systems it may be expected that the general appearance of the PLE spectra is similar. Comparison to our PLE spectra suggests that the dominant peaks in Fig. 2 are allowed heavy-hole transitions. This assignment is confirmed by a theoretical analysis.

Numerous calculations are made to fit the experimental peak positions. Initial fits are based on the $In_{0.53}Ga_{0.47}As$ bulk masses[19] $m_e^*=0.0410m_0$, $m_{hh}^*=0.4650m_0$, $m_{lh}^*=0.0503m_0$, and the band-gap discontinuity $\Delta E_g=635.0$ meV following from the InP FE peak position and the PL energy of the control layer.[20] Throughout these and all subsequent calculations we assume that no mass corrections are necessary for the heavy-hole states. In all cases, the finite square-well potential model is applied solving the associated transcendental equation relating energy eigenvalues E and potential depths V of the particles in the well. The initial calculations together with a comparison of the experimental spectra for different L_z indicate that large corrections of the high electron energies must be made irrespective of the conduction- (valence-) band offsets which are varied from $Q_e=25\%$ (i.e., $\Delta E_c=0.25\Delta E_g$) to $Q_e=65\%$. In all these calculations, the thicknesses of the layers as obtained from TEM analysis are treated as correct, and are varied only within their accuracy limits of approximately ± 5 Å.

Electron-mass corrections are applied following two different approaches, both theoretical and experimental. The theoretical masses of Yamada, Taguchi, and Sugimura,[21] calculated in the envelope-function approximation versus L_z, refer to the electron ground state. When these masses are applied to the excited states the resulting transition energies are too large to fit these spectra. The experimental masses of Portal *et al.*[22] were obtained from cyclotron resonance and magnetophonon resonance for QW's of $L_z = 80$, 100, and 150 Å and also refer to the electron ground states. We interpolate these masses by a relation $m_e(L_z)$ based on a third-order polynomial. Assume that the physical reason for the increase of m_e^* at smaller L_z is the increase of the electron energy subject to nonparabolicities (NP's) of the conduction band and independent of the quantum number n. Under this assumption the experimental excited electron states are compared to the $m_e^*(L_z)$ relation of Portal *et al.* reformulated as $m_e^*(E)$ by virtue of the experimental electron energies and the finite square-well potential model: Strong inconsistencies result for this comparison between wells of different widths. Again, these inconsistencies exist for band offsets Q_e varied from 36% to 60%. Application of the boundary conditions of Bastard[23] with an electron mass of $m_e^* = 0.079m_0$ for the InP barriers[24] also fails to produce acceptable fits to the spectra.

In contrast, reasonably good fits are obtained when conduction-band NP's are taken into account. Similar to recent work on GaAs-$Al_xGa_{1-x}As$ QW's[25] we represent NP's in terms of a parameter γ_e in the electron dispersion relation

$$E_e = (\hbar^2 k^2/2m_e^*)(1-\gamma_e k^2)$$

and use energy-dependent masses derived from this dispersion as

$$m_e(E_e)/m_0 = [-2+3(1-8\gamma_e m_0 E_e/\hbar^2)^{1/2}]^{-1} \ .$$

The fits shown in Fig. 2 are obtained with $\gamma_e = 3.3\times 10^{-15}$ cm^2 and $Q_e = 60\%$. In all of our attempts to fit the spectra the light-hole mass turns out to be too small; the fit in Fig. 2 uses $m_{lh} = 0.0850m_0$. We were unable to find a good fit assuming $Q_e = 36\%$, for L_z varied within ± 5 Å of the TEM values and γ_e and m_{lh} as free parameters. Our resulting band offset $Q_e \approx 60\%$ is inconsistent with $Q_e = 36\%$ derived from current-voltage and capacitance-voltage measurements on n-type $In_{0.53}Ga_{0.47}As$-n-type InP heterobarrier diodes,[26] and with $Q_e = 39\%$ from capacitance-voltage analysis of $In_{0.53}Ga_{0.47}As_yP_{1-y}$ heterojunctions lattice matched to InP for the entire compositional range from $In_{0.53}Ga_{0.47}As$ to InGaP.[27,28] It is consistent with $Q_e > 50\%$ estimated from the much less structured absorption spectrum of a 50-period $In_{0.53}Ga_{0.47}As$ sample with well thicknesses of $L_z = 125$ Å.[10]

In Fig. 3, we compare PL energy shifts, as a function of well-thickness L_z, with data from other works. Our experimental points lie substantially higher than the points compared, and are in good agreement with the three calculated curves displayed. Although the origin of the discrepancies between experimental upshifts and theory in the previous works is not known the agreement in the present case gives evidence for the superior quality of the samples grown by CBE. Differences between our points and our theoretical curve are mainly caused by the Stokes shifts between PL and PLE peaks which increase for smaller L_z. The relatively small differences between our calculated curve (dashed line in Fig. 3) and the other two curves (full lines) result from the assumption of different conduction or valence band offsets, respectively, as shown in Fig. 3 and different boundary conditions: The full lines refer to the Bastard model[23] assuming conservation of current, whereas we have neglected effects due to mass difference in the well and barrier material. In all three cases the electron and hole masses in $In_{0.53}Ga_{0.47}As$ are the same, with the energy dependent NP corrections of the electron mass for the dashed curve as discussed above. The near coincidence of the three calculated curves shows that the PL energy positions are not sensitive to the mass values and band offsets chosen as expected for the $n = 1$ electron-heavy-hole transitions.

In conclusion, we have measured excited states of well-confined excitons in $In_{0.53}Ga_{0.47}As$-InP quantum wells by means of PLE for a wide variety of well-widths L_z. We have assigned the states including a theoretical analysis yielding values for the band offsets and relevant electron and hole masses. This parameter set may not be unique and is subject to change with additional data. However, we state that the experimental data are not consistent with $Q_e \approx 40\%$, i.e., $\Delta E_c \approx 0.40$ (ΔE_g), and the conventional bulk masses; in contrast, it is consistent with $Q_e \approx 60\%$ and modified masses including essential nonparabolicity corrections for the electrons similar to the case of GaAs-AlGaAs quantum wells.

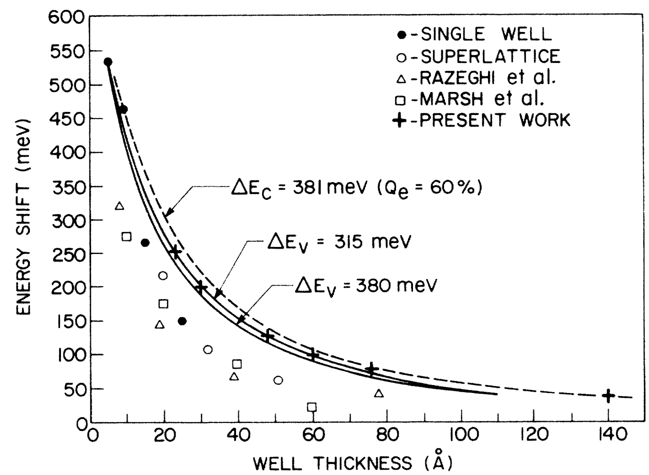

FIG. 3. Experimental and theoretical energy shifts from the $In_{0.53}Ga_{0.47}As$ band edge and comparison with data from other works (redrawn from Panish, Temkin, Hamm, and Chu, Ref. 29). The dashed curve, $\Delta E_c = 381$ meV, is from our data and uses the well thicknesses from the best fit to the PLE spectra. Razeghi and Duchemin refers to Ref. 30, Marsh, Roberts, and Claxton to Ref. 9.

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