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Luttinger parameters for GaAs determined from the intersubband transitions in GaAs/$Al_xGa_{1-x}As$ multiple quantum wells

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Electronic Raman scattering (ERS) measurements of photoexcited holes have been performed on GaAs/$Al_xGa_{1-x}As$ multiple quantum wells grown in the [111] *b* and [100] directions. These measurements indicate that the heavy- and light-hole masses along the [111] direction are $0.75m_0$ and $0.082m_0$, respectively. These values, compared with the heavy- and light-hole masses in the [100] direction ($0.34m_0$ and $0.094m_0$ respectively), reveal the highly anisotropic nature of the valence band in GaAs. We propose a new set of Luttinger parameters that describes this anisotropy.

As a result of the anisotropy of the band structure in bulk GaAs, one expects that the properties of GaAs/$Al_xGa_{1-x}As$ multiple quantum wells (MQW's) will depend on the relative orientation of the growth and crystallographic axes. Because of the recent advances in molecular-beam-epitaxial (MBE) growth,[1] this additional flexibility in "band-structure engineering" can now be employed in basic and applied studies of the electronic and optical properties of MQW's. We report the observation of intersubband transitions of photoexcited holes in undoped MQW's grown in the [111] *b* and [100] directions with resonant electronic Raman scattering (ERS). These measurements indicate that the valence band of bulk GaAs is highly anisotropic. We propose a new set of Luttinger parameters (LP) that describe this anisotropy and are consistent with the observed interband and intersubband excitations of [111] *b* and [100] MQW's. An adequate description of the in-plane dispersion of the valence subbands in GaAs/$Al_xGa_{1-x}As$ microstructures, a current topic of experimental[2,3] and theoretical[4-6] interest, requires accurate values for the LP.

The MQW samples examined in this study were undoped with barrier widths of 200 Å and alloy composition of $Al_{0.3}Ga_{0.7}As$. Typical samples were composed of 30 periods consisting of a GaAs well and an $Al_{0.3}Ga_{0.7}As$ barrier. The well widths estimated during growth are given by L_g in Table I. From the characteristics of the MBE machine, we estimate that L_g is accurate to within $\pm 10\%$ and that the reproducibility in the growth rate is better than $\pm 2\%$.

ERS measurements were performed in backscattering Raman geometries that are sensitive to spin-density and charge-density excitations.[7,8] In these configurations, we observe that the intersubband transitions occur at the same energies. Furthermore, the energies of the transitions are insensitive to laser intensity. Therefore, shifts in the intersubband transitions arising from either excitonic or depolarization phenomena[8] are expected to be small, and the measured transition energies should not differ greatly from the bare single particle intersubband transi-

TABLE I. Compilation of the characteristics of the [111] *b* MQW's examined in this study. L_g, L_R, and L_p are the quantum well widths estimated from the growth characteristics, intersubband transitions of photoexcited electrons, and photoreflectance spectroscopy. The heavy, M_{HH}, and light, M_{LH}, hole masses were determined by a comparison of the intersubband transitions of photoexcited holes with a calculation employing L_p for the quantum well width. The alloy composition of the barriers in these samples is $Al_{0.3}Ga_{0.7}As$.

L_g	L_R	L_p	M_{HH}	M_{LH}
400	360	370	...	...
300	285	287	...	...
250	...	238	0.75	0.082
200	...	198	0.70	...
150	...	145	0.75	...
100	...	93	0.80	0.082

tion energies.

Shown in Fig. 1 is the resonant electronic Raman scattering spectrum obtained at 10 K from a [111] *b* MQW with 198-Å-wide wells with an excitation energy of 1.611 eV. This spectrum, which was obtained at the peak of the resonant enhancement profile is characterized by two features labeled by E_{13} and E_{23}. The magnitudes of the shifts in the energies of these features as the width of the quantum well is changed and the sensitivity of the normalized intensity of these features to changes in laser power lead us to attribute these peaks to intersubband transitions of photoexcited holes. These processes are denoted schematically in the inset of Fig. 1. The resonant enhancement curves of the intersubband transitions are compared with the photoluminescence excitation (PLE) spectrum in Fig. 2. Absorption from the exciton associated with the third conduction subband and third heavy-hole subband, labeled by 33*H* in Fig. 2, gives rise to the peak in the photoluminescence excitation spectrum. The coincidence of peak positions of the 33*H* exciton and the resonance enhancement curves at 1.611 eV indicate that the 33*H* exciton is important in the ERS process. It is interesting to note that the final state of the intersubband transitions and the hole state of the 33*H* exciton involved in the resonance process are associated with the third heavy-hole subband. This observation is consistent with the prediction of Burstein *et al.*[9,10] that the two-step carrier density process is the dominant scattering mechanism for spin-density excitations.

Another interesting feature of the Raman measurement is illustrated in Fig. 3, where small increases in the energy of the intersubband transitions are observed as the excitation energy is increased. We propose that these shifts are a consequence of the inhomogeneous broadening of the 33*H* excitonic absorption arising from well width fluctuations.[11,12] Qualitatively, the low-(high) energy tail of the 33*H* excitonic absorption arises from quantum wells with a well width that is larger (smaller) than the average. This larger (smaller) well width results in a smaller (larger) intersubband transition energy. In order to test this proposition we have modeled the resonant enhancement factors of the Raman cross section described in Ref. 10 and, in addition, have added an inhomogeneous

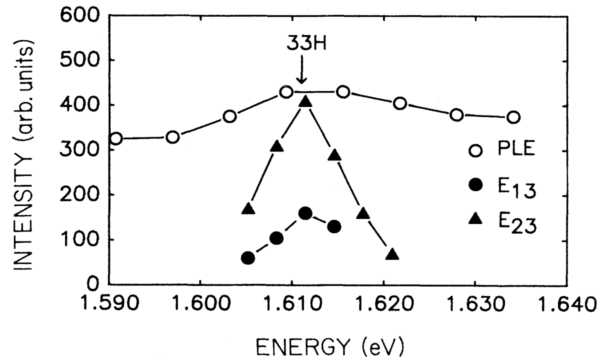

FIG. 2. A comparison between the resonance profile curves of the E_{13} and E_{23} intersubband transitions and the photoluminescence excitation (PLE) profile. The 33*H* label specifies the position of the interband exciton associated with the third heavy hole and third conduction subbands.

broadening of the excitonic transitions arising from well width fluctuations. This procedure produces resonance profiles and transition energies that are in semiquantitative agreement with those shown in Figs. 2 and 3.

In addition to the excitations shown in the Raman spectra presented in Fig. 1, we have observed eight additional intersubband transitions involving photoexcited holes from three other MQW's grown in the [111] *b* direction. The energies of these transitions are related to the heavy- and light-hole masses in the [111] direction. However, an accurate determination of the masses in this direction requires an accurate estimate of the quantum well width. Shown in Table I is a comparison between the magnitudes of the [111] *b* quantum well widths estimated from the growth conditions, L_g, and those estimated from electronic Raman scattering of photoexcited *electrons*, L_R, and our previously reported photoreflectance measurements, L_p.[13] Because the magnitude of the conduction-band mass is well established to be 0.0665, the energies of the intersubband transitions of photoexcited electrons allows the width of the quantum well to be determined accurately. The excellent agreement between L_R and L_p suggests that the well width estimated from photoreflectance is also fairly accurate. This is not surprising because the ener-

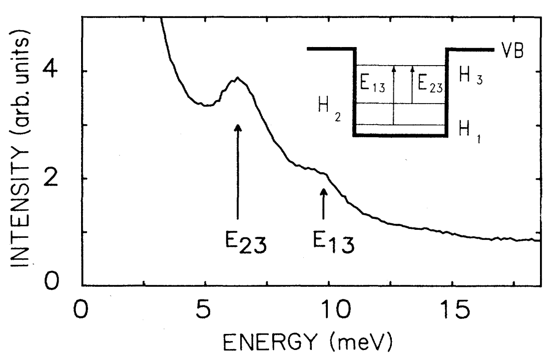

FIG. 1. Electronic Raman scattering spectrum of a [111] *b* MQW with $L_p = 198$ Å. The excitation energy is 1.611 eV. Shown in the inset are intersubband transitions E_{13} and E_{23} between the heavy-hole subbands labeled H_n.

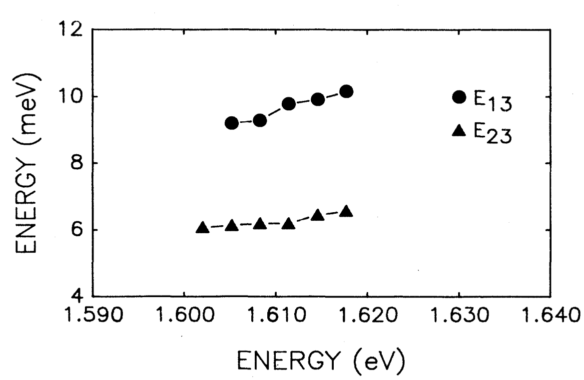

FIG. 3. An example of how the intersubband transitions shift as a function of resonant excitation energy.

gies of the excitonic transitions of [111] MQW's are mainly determined by the electron mass. We have estimated the heavy- and light-hole masses in the [111] direction by employing an isolated square-well model including current conserving boundary conditions.[14] The well width employed in the calculation was L_p and the intersubband transition energies at the peak of the enhancement curves were compared with the results of the calculation. The heavy- and light-hole masses that resulted in good agreement with the experiment on the [111] MQW's are shown in Table I. From the scatter in the heavy-hole masses listed in Table I for the four samples, we conclude that the heavy-hole mass in the [111] direction is $0.75 \pm 0.05 m_0$, respectively. Because of the large effective mass of the heavy holes, the intersubband transition energies obtained from the calculation are not significantly affected by the small changes in the valence-band offset. However, we estimate that the uncertainty in the light-hole mass in the [111] direction is $0.082 \pm 0.005 m_0$, where the errors in the determination of the mass are due to uncertainty in the magnitude of the valence-band offset. Valence-band offsets in the range of 0.55 (Ref. 13) to 0.68 (Ref. 15) were employed in the calculation.

A similar ERS study, although limited to two samples and five intersubband transitions, of [100] MQW's resulted in the heavy- and light-hole masses that are listed in Table II and are in good agreement with those previously proposed by Miller.[15,16]

Recently, other workers have reported interband absorption measurements on [111] MQW's that suggest that the heavy-hole mass in the [111] direction is near $0.9m_0$.[17] We believe that our determination of the masses is more accurate because intersubband transitions of holes are more sensitive to changes in the heavy-hole mass than are interband excitonic energies. As an illustrative example of this point, we consider the properties of a 100-Å-wide quantum well. If we change the heavy-hole mass from $0.9m_0$ to $0.8m_0$, the E_{13} intersubband transition changes from 27.3 to 30.37 meV (a difference of 3.07 meV), while the energy of the $11H$ exciton changes from 1.5415 to 1.5419 eV (a difference of 0.4 meV). Furthermore, because the energies of the excitonic transitions are determined mainly by the small conduction-band effective mass and the quantum well width, the width of the quantum well must be known to a high degree of accuracy. Finally, we note that experimental measurements of the binding energies for excitons in [111] MQW's are only beginning to be performed. Therefore, before interband measurements of [111] MQW's can be employed for an accurate determination of the valence masses, the exciton binding energies and widths of the quantum wells must be known to a high degree of accuracy.

The heavy- and light-hole masses, M_{HH} and M_{LH}, along the [100] and [111] directions are related to the Luttinger parameters through the following expressions:

$$M_{\mathrm{HH}} = (\gamma_1 - 2\gamma_2)^{-1}, \quad M_{\mathrm{LH}} = (\gamma_1 + 2\gamma_2)^{-1}, \quad [100],$$
$$M_{\mathrm{HH}} = (\gamma_1 - 2\gamma_3)^{-1}, \quad M_{\mathrm{LH}} = (\gamma_1 + 2\gamma_3)^{-1}, \quad [111]. \tag{1}$$

The dispersion curves for the valence bands of GaAs as a function of the Luttinger parameters are given by[18]

$$\varepsilon_{h,l} = -\tfrac{1}{2}\gamma_1 k^2 \pm [\gamma_2^2 k^4 + 3(\gamma_3^2 - \gamma_2^2) \times (k_x^2 k_z^2 + k_x^2 k_y^2 + k_y^2 k_z^2)]^{1/2}, \tag{2}$$

where γ_1, γ_2, and γ_3 are the Luttinger parameters and ε_h and ε_l are the energies of heavy- and light-hole valence bands. Table II presents a comparison between the heavy- and light-hole masses arising from Luttinger parameters reported by others[15,17,19–22] and those determined in the present study on MQW's grown in the [100] and [111] crystallographic directions.

An examination of Table II indicates that the Luttinger parameters recently proposed by Baldereschi and Binggeli result in heavy- and light-hole masses that are in the best agreement with our experimental studies of MQW's grown in the [111] and [100] directions. However, in light of the remaining disagreement between the masses determined from the studies of MQW's and those ascertained from Luttinger parameters proposed by others, we put forth a new set of Luttinger parameters based on the electronic Raman scattering studies of [100] and [111]

TABLE II. Listed in the upper four rows are Luttinger parameters that have been proposed previously by others. Also indicated are the corresponding heavy- and light-hole masses in the [111] and [100] directions. The heavy- and light-hole masses determined from measurements on [100] and [111] MQW's and the corresponding Luttinger parameters are given in the lower four rows.

				[100]		[111]	
Reference	γ_1	γ_2	γ_3	M_{HH}	M_{LH}	M_{HH}	M_{LH}
Lawaetz[a]	7.65	2.41	3.28	0.35	0.080	0.92	0.070
Skolnick[b]	6.98	2.25	2.88	0.40	0.087	0.82	0.078
Hess[c]	6.85	2.10	2.90	0.38	0.090	0.95	0.070
Baldereschi[d]	7.15	2.03	2.96	0.32	0.089	0.81	0.076
Hayakawa[e]	4.8	· · ·	1.85	· · ·	· · ·	0.90	0.117
Hayakawa[e]	5.74	1.39	· · ·	0.34	0.117	· · ·	· · ·
Miller[f]	6.8	1.9	· · ·	0.34	0.094	· · ·	· · ·
Proposed	6.8	1.9	2.73	0.34	0.094	0.75	0.082

[a] Reference 19.
[b] Reference 20.
[c] Reference 21.
[d] Reference 22.
[e] Reference 17.
[f] Reference 15.

MQW's presented here. Specifically, when Eq. (2) is combined with the masses determined from [111] and [100] MQW's, a new set of Luttinger parameters shown in Table II can be determined.

We have also performed several other experiments that test the accuracy of the LP that we are proposing. First, luminescence measurements were performed on [100] and [111] MQW's. In the case of [100] MQW's, when the energies of the excitonic transitions were combined with the known exciton binding energies[23] and the proposed LP's, the width of the quantum well could be calculated. This calculated well width was in good agreement with that implied by ERS measurements of intersubband transitions of photoexcited electrons performed on the samples. In the case of the [111] MQW's, we have estimated the exciton binding energies for the lowest heavy- and light-hole excitons by subtracting the measured excitonic energies from the calculated band to band transition energies. We observed excellent agreement between these binding energies and those predicted with the 4×4 Luttinger Hamiltonian as a function of quantum well width for MQW's grown in the [111] direction.[6]

In conclusion, we have performed intersubband transitions of photoexcited holes in [111] and [100] MQW's. This study suggests that the valence band of GaAs is highly anisotropic and that the heavy-hole mass in the [111] direction is about 2.2 times larger than that in the [100] direction. We propose a new set of Luttinger parameters for GaAs that describes this anisotropy and is consistent with the intersubband and interband transitions observed in [111] and [100] MQW's.

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