Energy-gap discontinuities and effective masses for GaAs-$Al_xGa_{1-x}As$ quantum wells

R. C. Miller, D. A. Kleinman, and A. C. Gossard
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 26 March 1984)

For the first time a single set of quantum-well parameters that describes the energies of the observed exciton transitions has been determined for both parabolic and square GaAs quantum wells grown with the GaAs-$Al_xGa_{1-x}As$ system. With the new set of parameters one can fit both the energies of the allowed $(\Delta n=0)$ heavy- and light-hole transitions, and the forbidden $(\Delta n\neq 0)$ heavy-hole transitions, for both types of quantum wells. For square wells, the GaAs well widths range from 51 to 521 Å. The new parameters include a conduction-band energy-gap discontinuity equal to $\sim 57\%$ of the total gap discontinuity and the following effective masses: $m_e^*/m_0=0.0665$, $m_h^*/m_0=0.34$, and $m_l/m_0=0.094$.

In contrast to a large body of earlier work,[1–4] a recent publication[5] on parabolic quantum wells grown with the GaAs-$Al_xGa_{1-x}As$ system presented evidence for a nearly equal division of the GaAs-$Al_xGa_{1-x}As$ energy-gap discontinuity ΔE_g between conduction and valence bands. The lineup of the bands at the heterointerface determines the potential barriers for the electrons and holes, which is of interest from a physics point of view and of importance for applications based on this system. Earlier papers on various aspects of square GaAs quantum wells,[1] and other structures,[2–4] obtained results consistent with the conventional masses $m_e^*/m_0=0.0665$ (Ref. 6) and $m_h^*/m_0=0.45$ (Ref. 7) and conduction- and valence-band discontinuities, $\Delta E_c=Q_e\Delta E_g$ and $\Delta E_v=Q_h\Delta E_g$, respectively, given by $Q_e=0.85=1-Q_h$. We refer to these as the "usual" parameters. For parabolic wells, however, a value $Q_e\sim 0.5$ was determined assuming the conventional masses. The value $Q_e\sim 0.5$ can be obtained from the relation for parabolic wells with infinite barriers[5]

$$\left(\frac{Q_e}{1-Q_e}\frac{m_h}{m_e^*}\right)^{1/2}=\frac{\Delta E_e}{\Delta E_h} \quad . \tag{1}$$

In Eq. (1) ΔE_e and ΔE_h are the electron and heavy-hole harmonic oscillatorlike energy-level spacings, respectively, and their ratio $\Delta E_e/\Delta E_h$ is observed to be 2.6 (Ref. 5) which therefore leads to $Q_e\sim 0.5$. At this point it should be noted that the parabolic well $Q_e\sim 0.5$ with the m^*'s cited above does not lead to satisfactory fits of observed series of exciton transitions for square wells, and conversely, $Q_e=0.85$ does not lead to acceptable fits of the parabolic-well data.

However, it is not generally appreciated that the usual square-well parameters do not give an adequate fit to certain aspects of the square-well data. In particular, it has been recognized for years,[1,8] but not emphasized, that a serious difficulty exists between calculations and experiment in connection with the transition energies of the square-well parity allowed "forbidden" (or "nondiagonal") transitions $(\Delta n\neq 0)$,[1,9] e.g., the transition involving the $n=1$ electron and $n=3$ heavy hole, denoted E_{13h}. For GaAs wells with $L=100$ Å and $x=0.30$ alloy barriers the calculated (usual parameters) and observed E_{13h} transition energies measured from the $n=1$ heavy-hole transition E_{1h} are 38 and 64 meV, respectively. In contrast, for parabolic wells, $Q_e\sim 0.5$ led to an excellent fit of the series of 11 observed diagonal $(\Delta n=0)$ and nondiagonal $(\Delta n\neq 0)$ heavy-hole exciton transitions.[5] This Rapid Communication addresses these discrepancies and proposes a new set of quantum-well parameters that lead to excellent calculated fits of both square and parabolic GaAs quantum-well data.

It is clear that what is required to fit the square-well forbidden transitions is a smaller heavy-hole mass and/or a larger Q_h (smaller Q_e) than usually assumed. In the present work the GaAs heavy-hole mass proposed is $m_h^*/m_0=0.34$, a value consistent with that utilized in some theoretical work[10,11] and also obtained in some earlier attempts to fit square-quantum-well data for wide GaAs wells.[12] With this m_h^* and the usual m_e^*, Eq. (1) leads to $Q_e=0.57$. Also to attempt a fit of the light-hole exciton data at the same time, use is made of a relation arising from the parabolic-well work[5]

$$\left(\frac{m_h}{m_l^*}\right)^{1/2}=\frac{\Delta E_l}{\Delta E_h}=1.9 \quad , \tag{2}$$

where ΔE_l is the observed light-hole exciton energy-level spacing. Equation (2) then yields $m_l^*/m_0=0.094$, a value about 7% larger than that (0.088, Ref. 6) preferred previously for square wells. Thus, the new parameters proposed for both parabolic and square GaAs quantum wells are $Q_e=0.57$, $m_e^*/m_0=0.0665$, $m_h^*/m_0=0.34$, and $m_l^*/m_0=0.094$.

The energies of the various light- and heavy-hole exciton transitions have been calculated with these new parameters as a function of L for square wells. The computation uses different effective masses for the particles in the well and barriers and the appropriate current-conserving boundary conditions for the wave functions at the heterointerfaces.[13] Effective masses for the alloy barriers are obtained from a linear extrapolation as a function of alloy composition x from the GaAs masses to the AlAs masses.[10,14] The logarithms of these calculated energies measured from the $n=1$ heavy-hole exciton transition E_{1h} vs L are shown in Fig. 1 as solid lines for $x=0.30$ alloy barriers. The exciton binding energy is taken to be a constant for the transitions in a given series, i.e., for either the heavy- or light-hole series, but the difference between the binding energies of the light- and heavy-hole excitons as a function of L (Refs. 15 and 16) is included in obtaining the curves in Fig. 1 so that they can be compared directly with the corresponding data. Also shown in Fig. 1 are these same energies determined from excitation spectra for ten different high-quality multi-quantum-well samples grown by molecular-beam epitaxy

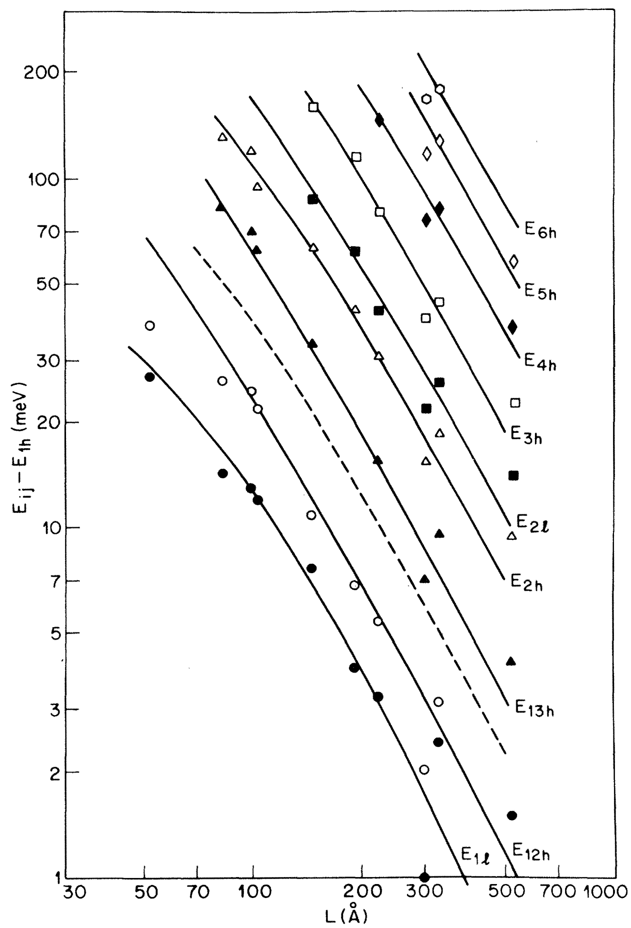

FIG. 1. Logarithm of the observed energies of the exciton transitions E_{ij} measured from the $n=1$ heavy-hole exciton E_{1h} vs logarithm of L. The solid lines are the calculated E_{ij}-E_{1h} using the new parameters $Q_e=0.57$, $m_e^*/m_0=0.0665$, $m_h^*/m_0=0.34$, and m_l^*/m_0 0.094. The dashed line is a calculated curve for E_{13h}-E_{1h} obtained with the usual parameters, $Q_e=0.85$ and $m_h^*/m_0=0.45$.

(MBE) with L ranging from 51 to 521 Å and alloy barriers with $x=0.30\pm0.05$. This uncertainty in x covers both the uncertainty in its determination and the actual range of the samples used for Fig. 1. It turns out that the calculated energies are not very sensitive to x. The fits of the calculated curves to the transitions are in general quite good.

For some samples, all the transitions for a given L are in excellent agreement with the calculated values as can be seen in Fig. 1. The relatively large spread in some of the data for the lower lying transitions is attributed to experimental uncertainties which are at best about ±0.4 meV and to effects due to strain which on a relative basis will affect these transitions the most.[17] In some cases, it appears that the sample L is incorrect, e.g., $L=300$ Å and $L=521$ Å. The well widths are estimated from the measured MBE growth rates and are in most cases believed accurate to about $\pm6\%$ at the point of measurement on the sample. However, for some samples, excellent fits of the observed energies of a series of exciton transitions to an extensive compilation of experimental data like that shown in Fig. 1 can only be achieved if the L differs from the growth rate estimated value by more than 10%. This suggests that L is not uniform across the wafer which is sometimes known to be the case from the growth geometry and which can also be demonstrated experimentally by observing the dependence of the energies of the transitions on the position on the sample of the focused excitation laser source. For the sample with $L=300$ Å mentioned above, a change of L to 340 Å results in a mean deviation between the shifted data points and the calculated curves for the eight transitions of 1.0 meV. It should be noted that for $L \gtrsim 200$ Å there is on occasion some weak structure present in the excitation spectra, from which the data in Fig. 1 are derived, which has not been identified with known exciton transitions or other mechanisms.

For a comparison with the usual square-well parameters, the calculated curve for the E_{13h} forbidden transitions obtained with the usual parameters is shown in Fig. 1 as a dashed line. The data are in poor agreement with the dashed line over the entire range of L. However, for higher lying transitions, i.e., $E_{ij}-E_{1h}$ for $i>2$, the differences between the calculated energies using the usual and new parameters is small compared to the spread in the experimental data. It is surprising indeed that $Q_e=0.85$, $m_h^*/m_0=0.45$, and $Q_e=0.57$, $m_h^*/m_0=0.34$ can result in very nearly the same transition energies. This emphasizes the importance of the forbidden transitions and the parabolic quantum wells for determining the GaAs quantum-well parameters.

It can be seen in Fig. 1 that the light-hole data are also fitted by the new set of parameters which include $m_l^*/m_0=0.094$. In addition, the calculated and observed values of the E_{1h} exciton transitions from which the energies to the E_{ij} transitions are plotted in Fig. 1 agree to within about 1 meV when the exciton binding energy[15,16] is included in the calculation.

The new parameters have been obtained in such a manner that Eqs. (1) and (2) are automatically satisfied for the parabolic-well samples. But in order to fit the observed magnitudes of the energy ladder spacings for the principal parabolic-well sample,[5] L is set equal to 540 Å. This value is within the range of the nominal value of $L=510\pm35$ Å estimated earlier[5] from the measured growth rates. With $L=540$ Å, the exact calculation of the exciton transitions taking into account finite wells, etc., as described in Ref. 5, yields values in excellent agreement with the data for both the diagonal and nondiagonal heavy-hole transitions, and for the light-hole transitions. A similar approach to the other two parabolic-well samples[5] also leads to good agreement between the calculations and experiment.

The calculations shown in Fig. 1, and those for the parabolic wells, do not take into account the nonparabolicity (NP) of the conduction and light-hole bands. (For the heavy hole, NP is negligible.) We have investigated in detail the effect of the electron NP on the calculated energies. We represent NP in terms of a parameter γ_e in the equation for the band energy

$$E_e=(\hbar^2k^2/2m_e^*)(1-\gamma_e k^2) \quad . \tag{3}$$

We have calculated γ_e from the five-band model used by Lawaetz,[10] fitting the parameters of the model to the observed energy gaps and the masses $m_e^*=0.0665m_0$, $m_h^*=0.34m_0$, $m_l^*=0.094m_0$. In this way we obtain for GaAs the value $\gamma_e=4.9\times10^{-15}$ cm^2. This may be compared to the value $\gamma_e=3.1\times10^{-15}$ cm^2 obtained by Raymond, Robert, and Bernard[18] (expressed in our notation) from a three-band (Kane) model. Both values are in reasonable agreement with the experimental data cited in

Ref. 18. Using our value of γ_e, we have recalculated the curves shown in Fig. 1. We find that at the highest energies shown (> 100 meV) there is a downward shift of the curves of order 6%–8%, which would be barely resolvable in Fig. 1 and is too small to upset any of our conclusions. At lower energies the effects of NP are completely negligible. Similarly, NP for the light hole is unimportant here even though $\gamma_l \sim 1.5\gamma_e$.

For the parabolic wells we have calculated the expectation values of the NP perturbation in the Hamiltonian for the electron. Our result can be written

$$\langle n|H_{\rm NP}|n\rangle = -\tfrac{3}{8}(\gamma_e m_e^*/\hbar^2)(\Delta E_e)^2(2n^2-2n+3)\ , \quad (4)$$

$$n=1,2,\ldots\ .$$

From this we calculated that due to NP the spacing of the levels in the electron ladder diminishes with increasing n by amounts ranging from 1.4% for E_2-E_1 to 7.2% for E_6-E_5 (E_6 is the highest electron level assigned in Ref. 5). These shifts are smaller than experimental uncertainties, e.g., that due to broadening, so we conclude that NP has no effect on the conclusions reached here.

In summary, a single set of GaAs quantum-well parameters is proposed that leads to excellent agreement between the observed and calculated exciton transitions for both square and parabolic potential wells. These transitions include both the allowed heavy- and light-hole excitons, and the forbidden heavy-hole excitons. The new parameters are $\Delta E_c = 0.57\Delta E_g$, $m_e^*/m_0 = 0.0665$, $m_h^*/m_0 = 0.34$, and $m_l^*/m_0 = 0.094$. It is suspected that a "fine tuning" of these parameters could lead to slightly different values, but the extensive data cited herein, and other data,[19] lend strong support to $Q_e \sim 0.6$ in contrast to the previously accepted value $Q_e \sim 0.85$.

[1]R. Dingle, in *Festkörperprobleme,* edited by H. J. Queisser, Advances in Solid State Physics, Vol. 15 (Pergamon/Vieweg, Braunshweig, 1975), p. 21.
[2]A. C. Gossard, W. Brown, C. L. Allyn, and W. Wiegmann, J. Vac. Sci. Technol. 20, 694 (1982).
[3]J. Sanchez-Dehesa and C. Tejedor, Phys. Rev. B 26, 5824 (1982).
[4]R. People, K. W. Wecht, K. Alavi, and A. Y. Cho, Appl. Phys. Lett. 43, 118 (1983).
[5]R. C. Miller, A. C. Gossard, D. A. Kleinman, and O. Munteanu, Phys. Rev. B 29, 3740 (1984).
[6]Q. H. F. Vrehen, J. Phys. Chem. Solids 29, 129 (1968).
[7]A. L. Mears and R. A. Stradling, J. Phys. C 4, L22 (1971).
[8]R. C. Miller and D. A. Kleinman (unpublished).
[9]R. C. Miller, D. A. Kleinman, W. A. Nordland, Jr., and A. C. Gossard, Phys. Rev. B 22, 863 (1980).
[10]P. Lawaetz, Phys. Rev. B 4, 3460 (1971).
[11]A. Baldereschi and N. O. Lipari, Phys. Rev. B 3, 439 (1971).
[12]D. A. Kleinman, Phys. Rev. B 28, 871 (1983).
[13]G. Bastard, Phys. Rev. B 24, 5693 (1981).
[14]D. J. Stukel and R. N. Euwema, Phys. Rev. 188, 1193 (1969).
[15]R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B 24, 1134 (1981).
[16]R. L. Greene and K. K. Bajaj, Solid State Commun. 45, 831 (1983).
[17]R. Dingle and W. Wiegmann, J. Appl. Phys. 46, 4312 (1975).
[18]A. Raymond, J. L. Robert, and C. Bernard, J. Phys. C 12, 2289 (1979).
[19]A. C. Gossard (unpublished).