Reply to "Comment on 'Determination of valence-band discontinuity via optical transitions in ultrathin quantum wells' "

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The salient points of our previous paper [Phys. Rev. B 33, 7259 (1986)] are reinforced in this Reply to the preceding Comment by Miller.

The essential points of our work[1] were to demonstrate *what* experimental data from optical transitions should be used and *how sensitive* they are to the Q value of band offset. Contrary to Miller's comment,[2] Dingle *et al.*[3] did not emphasize how sensitive the Q value depended upon the value of the energy separation, $\Delta E(L_z)$, between the $n=1$ heavy- and light-hole subbands. We found[1] that the values of ΔE are most sensitive to the band offset for L_z ranging from 15 to 80 Å. Based on this key finding a currently fashionable choice of $Q_v=0.40$ proposed by Miller *et al.*[4] was ruled out using Dingle's connection rule and the then available experimental data in this range of L_z. Another key point made in our paper[1] was that systematic measurements should be performed in the sensitive zone of well width (15 Å to 80 Å) to precisely determine the Q value. These essential points are still overlooked in depth by Miller's comment[2] to our paper.[1]

It should be pointed out here that the extent of data used in our paper[1] was not the main issue of the paper and more data could not alter the essential spirit of the paper. Most recently, Miller *et al.*[5] have performed new measurements providing more data in the sensitive zone which will be discussed below in this Comment.

We have recognized that there has been a large body of data pointing to a larger $Q_v \sim 0.40$, which was the mainstream of thought in this field. However, most of the experimental probes carry their own source of uncertainties.[6] The electrical measurements are often plagued by residual doping whereas the intersubband optical transitions may prove to be too weakly dependent upon the band offset. The optical method described in our paper[1] provides a sensitive optical test of band offsets in $GaAs/Al_xGaAs_{1-x}$ structures. It should be mentioned that there are some most recent determinations of Q value for $GaAs/Al_xGa_{1-x}As$ which do not yield $Q_v=0.40$ but point to values of 0.31 from Raman scattering data[7] and 0.23 ± 0.07 from electrolyte electroreflectance study.[8]

As noticed by Miller,[2] one of the data in the original paper[1] to support lower Q_v was obtained in a circular fashion. However, the other data quoted by us do not suffer from this problem.

It is well known that the mismatch of effective mass in well and barrier is responsible for the appearance of various connection rules[3,9,10] for the envelope wave function and its derivative. In general, using a different connection rule will result in different eigen energy levels and hence differences in the spacing between the levels. Therefore, the Q value determined by ΔE versus L_z depends much on what connection rule is employed. This is where some of the problems come from.

In the spirit of our past paper, the light- and heavy-hole energy splitting is plotted in Fig. 1 as function of well thickness L_z using various connection rules.[3,9,11] The Q value is adjusted in such a way the ΔEs for $L_z > 40$ Å calculated[12] using different connection rules coincide with each other within ~ 2 meV. D, B, and N in the Fig. 1 refer to the Dingle's connection rule, Bastard's connection rule, and new connection rule.[11] The material parameters used for the calculation of the

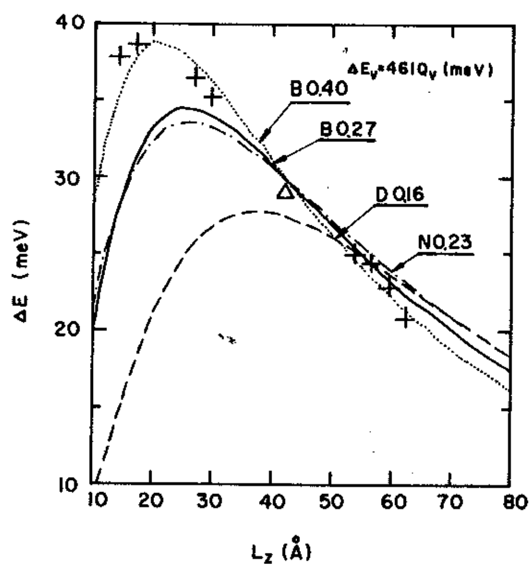

FIG. 1. Calculated light- and heavy-hole energy splitting as function of L_z. D, B, and N refer to Dingle's, Bastard's, and new connection rules. The values of Q_v are indicated on the corresponding curves. The masses used for the dotted curve is Miller's masses. The masses used for the other curves are the conventional masses given in the text. The plus and triangle data are from Refs. 5 and 15, respectively.

curves are $x=0.37$, $\Delta E_v = 1247xQ_v$ (meV) (the value of Q_v is indicated next to the corresponding variable in the Fig. 1), masses[13,14] of heavy hole (light hole) for GaAs and AlAs are $0.403m_0$ ($0.087m_0$) and $0.487m_0$ ($0.208m_0$), respectively; except for the curve where the heavy-hole mass ($0.34m_0$) and light-hole mass ($0.094m_0$) proposed by Miller *et al.*[4] were used. The plus and triangle data points in Fig. 1 are from Refs. 5 and 15, respectively.

Figure 1 shows two important features. First, as expected the energy splitting ΔE is sensitive to the connection rule in the sensitive zone of L_z, especially for $L_z < 40$ Å. This features makes it possible to experimentally demonstrate which connection rule is appropriate by systematically measuring ΔE in the sensitive zone. Based on the data given by Miller *et al.*,[5,15] Dingle's connection rule which was used by us to show how to determine Q value in the sensitive zone of L_z seems to be not appropriate. However, the method itself described in our paper[1] is still very useful. Second, for the given connection rule the splitting ΔE is very sensitive to the effective masses. In order to fit simultaneously the data given by Miller *et al.*[5,15] it seems to be necessary to use the Bastard's connection rule and the mass parameter set ($m_{\rm HH}=0.34m_0$, $m_{\rm LH}=0.094m_0$) and $Q_v=0.40$ proposed by Miller *et al.*[4] The use of new connection rule[11] with $Q_v=0.23$ seems to fit the data for $L_z > 30$ Å. It should be pointed out here that in our calculation the binding-energy difference between the light hole and the heavy hole is taken to be a constant (~ 0.5 meV). This may be a good approximation for $L_z > 40$ Å. In order to compare the measured ΔE with the calculated ΔE for $L_z < 40$ Å an exact analysis taking the proper exciton binding energy into account must be performed.[16]

In Fig. 2, the energy separation ΔE_{12} between first and second conduction subbands is plotted as function of L_z using three different connection rules by including[17] the energy-dependent mass $m_w(E_n)$ (Ref. 18) as well as $m_b(E_n)$, $n=1,2$, for well material GaAs and barrier material $\rm Al_{0.3}Ga_{0.7}As$, respectively, and compared ΔE_{12} with the published experimental data by West and Eglash.[19] This work is not affected by the valence-band complexity, the exciton binding energy, and can provide an independent test of Q value.

The $m_b(E_n)$ is obtained by

$$\frac{\partial^2}{\partial k^2}\left[\left[\frac{E_g^2}{4}+\frac{\hbar^2k^2E_g}{2m_{\rm CB}}\right]^{1/2}-\frac{E_g}{2}\right]=\frac{\partial^2}{\partial k^2}\left[\frac{\hbar^2k^2}{2m_b(E_n)}\right]. \tag{1}$$

The expression inside of the large brackets on the left-hand side of Eq. (1) is the energy-momentum relationship within semiconductor band gap.[20] After differentiation the $m_b(E_n)$ reduced to

$$m_b(E_n)=m_{\rm CB}(1+4\chi_n)^{3/2}, \quad n=1,2,\ldots, \tag{2}$$

where $m_{\rm CB}$ is electron effective mass at conduction band edge of $\rm Al_{0.3}Ga_{0.7}As$, $\chi_n = |\hbar^2k_{bn}^2/2m_{\rm CB}E_g|$, and $n=1,2$ for first and second subbands. Note that k_b^2 is negative and k_b is imaginary reflecting the electron wave function within the gap of $\rm Al_{0.3}Ga_{0.7}As$ is exponentially damped. The dispersion relations dependent upon the real and imaginary wave vectors in the well and the barrier are schematically shown in the inset of Fig. 2. The connection rule and the Q value used are indicated on the corresponding curves. The value of x is equal to 0.30 in order to make a comparison with the data ΔE_{12} of West and Eglash.[19]

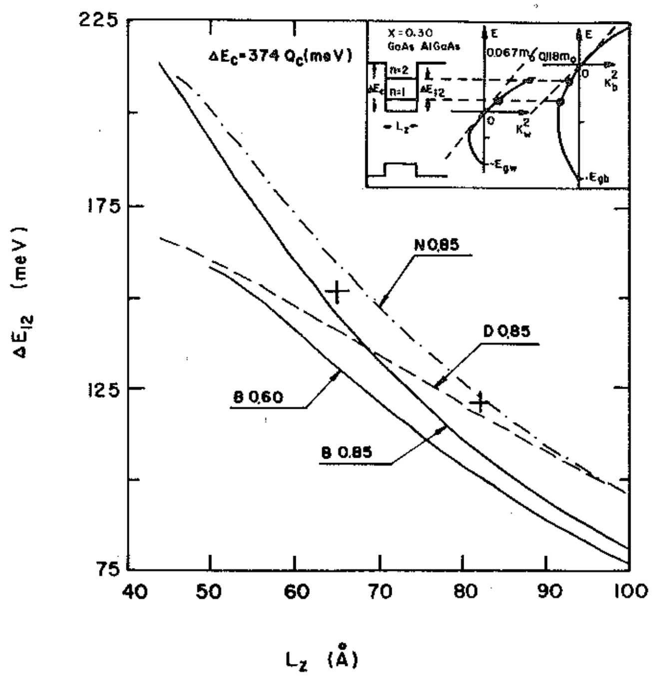

FIG. 2. Calculated energy separation ΔE_{12} between first and second conduction subbands as a function of well width L_z using Dingle's (D), Bastard's (B), and new (N) connection rules. The curves are calculated using $\Delta E_c = Q_c \Delta E_g$ ($\Delta E_g = 1247x$ meV, $x=0.30$). The values of Q_c are indicated on the corresponding curves. The data of West and Eglash (Ref. 19) are indicated as two plus signs. The inset shows schematically the well and the energy-momentum dispersion curves. The electron effective masses at band edges for GaAs and $\rm Al_{0.3}Ga_{0.7}As$ are also indicated in the inset.

Several features are very apparent in Fig. 2. First, for the given connection rule ΔE_{12} is very sensitive to the Q value in the sensitive zone of L_z giving rise to a possibility of extracting the value of ΔE_c and Q value. Second, the value of ΔE_{12} calculated using the new connection rule approaches the values of ΔE_{12} at ~ 45 Å and at $L_z > 100$ Å calculated using Bastard's and Dingle's connection rules, respectively. Third, the two experimental points support neither Bastard's nor Dingle's connection rules, but seems to be more favorable to the new connection rule with $Q_c = 0.77$.

In conclusion, we have shown different connection rules and hole masses yield different Q values. Furthermore, the appropriate connection rule can be discriminated by systematically measuring ΔE and ΔE_{12} in the sensitive zone of L_z. Dingle's 85-15 rule is based on his connection rule while Miller's 60-40 rule is based on Miller's effective masses and Bastard's connection rule.

The essential information described in our paper[1] is not compromised by the comments raised by Miller but reinforced here. We stress that more systematic photoluminescence, PLE, and inter-conduction-subband transition data must be taken in the sensitive zone (15 to 80 Å) to determine the proper connection rule and an accurate Q value.

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