Sharp-line photoluminescence spectra from GaAs-GaAlAs multiple-quantum-well structures

D. C. Reynolds, K. K. Bajaj, and C. W. Litton
Avionics Laboratory, Air Force Wright Aeronautical Laboratories (AFWAL/AADR), Wright-Patterson Air Force Base, Dayton, Ohio 45433

P. W. Yu
University Research Center, Wright State University, Dayton, Ohio 45435

W. T. Masselink
Department of Physics and Coordinated Science Laboratory, University of Illinois, Urbana, Illinois 61801

R. Fischer and H. Morkoc
Department of Electrical Engineering and Coordinated Science Laboratory, University of Illinois, Urbana, Illinois 61801
(Received 16 January 1984; revised manuscript received 12 April 1984)

The photoluminescence spectra from four $Al_{0.25}Ga_{0.75}As$-GaAs multiple-quantum-well (MQW) structures with well thicknesses of 100, 200, 300, and 400 Å and barrier thicknessess of 100 Å have been interpreted. The extrinsic emission is predominantly in the donor region. A number of very sharp transitions have been observed for the first time. Several of the transitions are no more than 0.15 meV in width. The light- and the heavy-hole free excitons were observed in the 400 Å well. The heavy-hole free excitons were observed in the other wells. The radiative transitions associated with the neutral-donor bound excitons as well as the free-to-bound and neutral-acceptor bound excitons are discussed. Magnetic field splittings were observed for several of the optical transitions. These data together with the diamagnetic shifts and excitation-dependent measurements are used in the identification of the multiple-quantum-well transitions.

INTRODUCTION

The interest in multiple-quantum-well (MQW) structures has been growing rapidly both from the standpoint of purely scientific investigations as well as the practical aspects of devices produced from these structures. Photoluminescence techniques have been used extensively to investigate the properties of the MQW structures. Considerable effort has been devoted to investigating the intrinsic character of these wells.[1–6] Extrinsic properties of the wells have, however, received less attention.[7,8] The light- and heavy-hole free-exciton energies as a function of well thickness (L_z) have been experimentally measured[5] and have also been calculated.[5,9–11]

The present investigation focuses on the interpretation of the photoluminescence from $Al_{0.25}Ga_{0.75}As$-GaAs MQW with L_z of 100, 200, 300, and 400 Å. The extrinsic emission is predominately in the donor region. A number of very sharp transitions have been observed. The sharpness of the transitions is a unique feature of this work, contrasting it with much of the published literature. Several of the transitions have full widths at half maximum (FWHM) of no more than 0.15 meV. These features reflect very high-quality MQW structures. Both the light- and heavy-hole free excitons were observed in the 400-Å well. In the thinner wells the light-hole free excitons were not observed; the heavy-hole free excitons were observed in all of the wells. The transitions believed to be associated with donor-bound–heavy-hole excitons, D^0X_h, were observed in all the wells, whereas those associated with donor-bound–light-hole excitons were observed only in 300- and 400-Å wells. Free-to-bound transitions, D^0h, (free hole to donor) were observed for both light and heavy holes. Finally, the acceptor-bound–heavy-hole exciton, A^0X_h transitions were observed in all of the wells.

Magnetic field splittings were observed for several of the optical transitions. These data together with diamagnetic shifts were used in identifying the MQW transitions. The MQW transitions have smaller diamagnetic shifts than their counterparts in bulk GaAs. This permits the identification of MQW transitions occurring in the region of bulk GaAs transitions. Also, excitation-intensity-dependent measurements were used to aid in the identification of MQW transitions.

GROWTH CONDITIONS

The structures reported here were grown on (100) oriented Si-doped n-type GaAs substrates by molecular-beam epitaxy. The substrates were prepared and loaded in the growth chamber as described in Ref. 6. The multiple-quantum-well structures were grown at 600 °C. A two-stage thermal cracker[6] oven was utilized to obtain As_2 which was used as an anion source throughout the growth. The excellent results reported here are believed to be, at least in part, a result of the use of dimeric arsenic obtained in this manner. The oven is capable of providing enough arsenic for about 700 μm of epitaxial growth.

EXPERIMENTAL METHOD

The experimental apparatus employed in this investigation permitted high-resolution photoluminescence measurements to be performed at 2 K and in magnetic fields up to 36 kG. In the intrinsic region of GaAs a dispersion of 0.54 Å/mm was achieved using a 4-m spectrometer. Data were collected on Kodak-type-1-N spectrographic plates. The photoluminescence was excited with the 6471-Å line of a krypton ion laser. Also, the emission spectra were analyzed by a 1.26-m Spex spectrometer for the excitation-intensity-dependent measurements. For this purpose a lock-in amplifier was used for the standard synchronous detection with a cooled RCA C31034 photomultiplier tube. The excitation intensities were in the range of 10^{-3}–10 W/cm^2.

EXPERIMENTAL RESULTS

The samples used in this study were MQW structures which consisted of $Ga_{0.75}Al_{0.25}As$ barrier layers of thickness 100 Å and GaAs well thickness of 100, 200, 300, and 400 Å. The total number of cycles in the 300- and 400-Å-thick MQW samples was 20, whereas the 200- and 100-MQW samples had 33 and 50 cycles, respectively. In general, the sharpest transitions are observed in those wells with the largest L_z; however, the heavy-hole D^0X transition in the 100-Å well was 0.2 meV FWHM. The photoluminescence for the 400-Å well is shown in Fig. 1 as this structure exhibits all the relevant transitions. In this sample both the light- and heavy-hole free excitons can be seen. Several extrinsic transitions are also observed; some of them occur in the same spectral region as some of the bulk GaAs transitions. The identification of the MQW transitions is aided by their diamagnetic shifts in a magnetic field. The GaAs transitions have larger diamagnetic shifts than the MQW transitions. It can be seen in Fig. 1 that the GaAs buffer layer is of very high quality. The D^0X transition and its first nonrigid rotational state are separated by just 50 μV, and are clearly resolved, thus reflecting the high quality of the layer. Both the light- and heavy-hole free excitons are appreciably less intense than the extrinsic transitions with the heavy-hole free exciton being the more intense. The MQW transitions are marked by arrows in Fig. 1 with the identification of each transition being listed. The behavior of some of the MQW transitions in an applied magnetic field is shown in Fig. 2. The sharpest lines in all of the MQW tansitions are the free exciton and the D^0X transitions. In the 400-Å well the light- and heavy-hole free excitons show a doublet splitting with the light-hole exciton having the greater effective g value. The D^0X_1 transition also shows a doublet splitting. The D^0X_h transition comes in a region where bulk GaAs emission is also observed making the resolution of this transition difficult. In the 300-Å well the light-hole free exciton is not seen. The heavy-hole free-exciton splitting could not be resolved, however, the splitting of the D^0X_1 is clearly resolved. If it can be assumed that the magnetic field splitting of the MQW free excitons can be patterned after those in bulk nondegenerate semiconductors then it would be expected that the light-hole free exciton will split as the sum of the electron and hole g values. The heavy-hole free excitons will split as the difference of the electron and hole g values. Analyzing the magnetic field splitting of the free excitons in the 400-Å well based on the above assumption and further assuming an electron g value of 0.5, essentially that of the free electron g value in bulk GaAs, one obtains a light-hole g value of 1.3 and a heavy-hole g value of 0.46. It would be expected that the D^0X transitions would show a four-line splitting. A more detailed analysis of the magnetic field splittings will require still sharper transitions. Both the splittings and the diamagnetic shifts of MQW transitions aid in the identification of these transitions. The excitation intensity was also varied to differentiate transitions in MQW

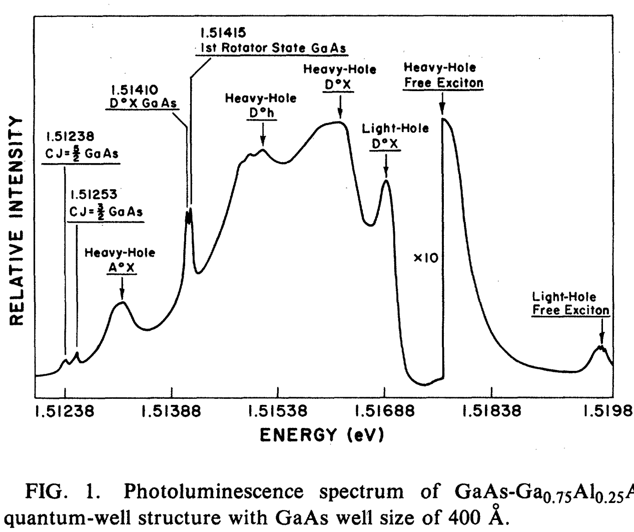

FIG. 1. Photoluminescence spectrum of GaAs-$Ga_{0.75}Al_{0.25}As$ quantum-well structure with GaAs well size of 400 Å.

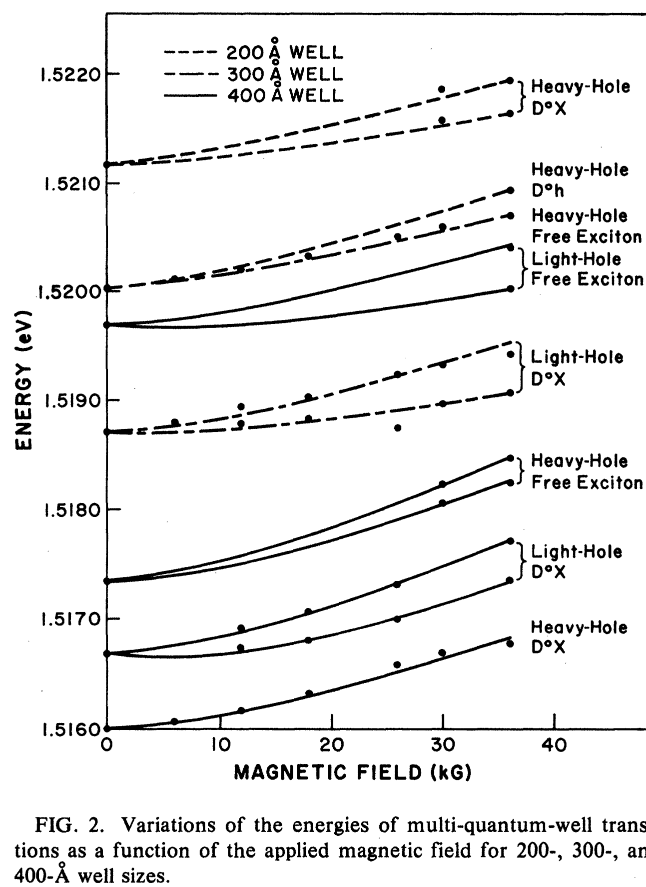

FIG. 2. Variations of the energies of multi-quantum-well transitions as a function of the applied magnetic field for 200-, 300-, and 400-Å well sizes.

and bulk GaAs buffer layers to identify the transitions in MQW. The emissions from the GaAs buffer layer decrease with the excitation intensity relatively faster than those from MQW. Thus, at a low excitation intensity, the emissions from MQW are primarily present with the quenching of the GaAs transitions shown in Fig. 1. The dominant MQW emissions in the excitation intensity range investigated are due to the D^0X_h and D^0h transitions. This is consistent with the observation made on bulk GaAs as a function of excitation intensity. The X_h, D^0X_1, and D^0X_h emission in MQW generally follow a linear emission-excitation relationship as expected.

The variation of the MQW transitions as a function of L_z is shown in Fig. 3. It is seen that the free and bound excitons extrapolate to the analogous transitions in bulk GaAs. The light-hole free exciton is only observed in the 400-Å well; the exciton energy is calculated for the other wells.[10] The free-to-bound transitions are all calculated from the following expression:

$$E_{D^0,h} = E_e - E_{D^0} + E_h + E_g ,$$

where E_{D^0h} is the energy of the free-to-bound transition, E_e is the electron subband energy, E_h is the hole subband energy, E_g is the band gap, and E_{D^0} is the neutral-donor binding energy. The energies E_e, E_h, and E_{D^0} are theoretical values from Greene and co-workers.[10,12] The appropriate values are taken for both the light- and heavy-hole exciton and are plotted as the X's in Fig. 3. The solid dots are the experimental values. The heavy-hole D^0h is observed for all of the wells; the light-hole D^0h transitions come close to the heavy-hole free-exciton transition for the 400- and 300-Å wells and occur on the high-energy side of the heavy-hole free exciton for smaller L_z. The remaining free-exciton and bound-exciton transitions are appropriately labeled in Fig. 3. Two transitions have been observed in the 100-Å well for which assignments have not yet been made. We find that our values of the heavy-hole D^0h and light-hole D^0h transitions agree rather well with those measured by Lambert, Deveaud, Regreny, and Talalaeff.[8] In addition, the behavior of A^0X_h transition as a function of the well size is very similar to that found by Miller, Gossard, Tsang, and Munteanu,[7] namely, the dissociation energy of the exciton increases as the well size is reduced. Recently, Kleinman[13] has calculated the dissociation energy of an exciton bound to a neutral donor as a function of the quantum-well size. He used a variational approach assuming infinite potential barriers. He finds that the dissociation energy increases as the well size is reduced. The dissociation energy of the heavy-hole exciton obtained by subtracting the energy of the D^0h transition (Fig. 3, curve e) from that of the X_h transition (Fig. 3, curve c) increases as L is reduced. The increase is somewhat smaller than that predicted by Kleinman.[13] The measured value of the dissociation energy, however, for a given value of the quantum-well size, is larger than the variationally calculated value.

It should be pointed out that in the interpretation of our data we have assumed that the donor impurity ion is located at the center of the well. It is known[14,15] that the binding energy of a donor depends on its location in the well. For donors located at the interface the binding energy is smaller by as much as 5 meV. The density of states per unit binding energy has a maximum for donors at the center and a smaller maximum for donors at the interface.[14,16] The strongest transitions are therefore those associated with the donors at the center. We believe that the transitions we observe are associated with donors located at or near the center of the well.

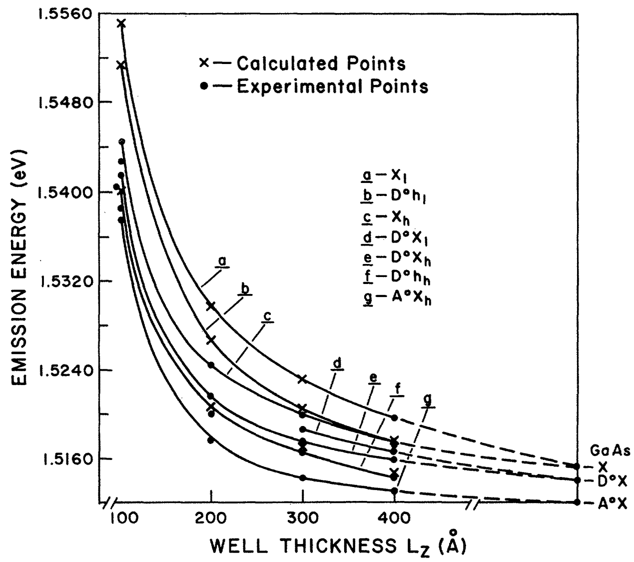

FIG. 3. Variations of the emission energies of free light-hole exciton (X_l), free heavy-hole exciton (X_h), light-hole exciton bound to a neutral donor (D^0X_l), heavy-hole exciton bound to a neutral donor (D^0,X_h), heavy-hole exciton bound to a neutral acceptor (A^0X_h), neutral-donor–light-hole (D^0h_l), and neutral-donor–heavy-hole (D^0h_h) transitions as a function of GaAs well thickness (L_z).

CONCLUSIONS

Very sharp (0.15 meV FWHM) optical transitions have been observed in MQW structures. The very narrow linewidths are unique among published MQW transitions, reflecting the very high quality of these structures. These results suggest that even sharper transitions will be achieved in the future. This will greatly assist in the identification and analysis of MQW transitions. In this investigation both light- and heavy-hole free excitons were observed, as well as the D^0X_1 and D^0X_h transitions. Free-to-bound D^0h and A^0X_h were also identified. The transitions were tracked through MQW structures as L_z was varied. Magnetic field data along with excitation-intensity-dependent measurements were used to assist in the identification of the transitions.

[1]L. Esaki and R. Tsu, IBM J. Res. Dev. 14, 61 (1970).

[2]R. Dingle, *Festkoerperprobleme*, edited by H. J. Queisser, Advances in Solid State Physics, Vol 15 (Pergamon/Vieweg, Braunschweig, 1975), p. 21.

[3]B. A. Vojak, N. Holonyak, Jr., D. W. Laidig, K. Hess, J. J. Coleman, and P. D. Dapkus, Solid State Commun. 35, 477 (1980).

[4]R. C. Miller, D. A. Kleinman, W. A. Nordland, Jr., and A. C. Gossard, Phys. Rev. B 22, 863 (1980).

[5] R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B 24, 1134 (1981).
[6] W. T. Masselink, Y. L. Sun, R. Fischer, T. J. Drummond, Y. C. Chang, M. V. Klein, and H. Morkoc, Appl. Phys. Lett. 44, 435 (1984).
[7] R. C. Miller, A. C. Gossard, W. T. Tsang, and O. Munteanu, Solid State Commun. 43, 519 (1982); Phys. Rev. B 25, 3871 (1982).
[8] B. Lambert, B. Deveaud, A. Regreny, and G. Talalaeff, Solid State Commun. 43, 443 (1982).
[9] G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Phys. Rev. B 26, 1974 (1982).
[10] Ronald L. Greene and K. K. Bajaj, Solid State Commun. 45, 831 (1983).
[11] Ronald L. Greene, K. K. Bajaj, and Dwight E. Phelps, Phys. Rev. B 29, 1807 (1984).
[12] Ronald L. Greene and K. K. Bajaj, Solid State Commun. 45, 825 (1983).
[13] D. A. Kleinman, Phys. Rev. B 28, 871 (1983).
[14] G. Bastard, Phys. Rev. B 24, 4714 (1981).
[15] C. Mailhiot, Y. C. Chang, and T. C. McGill, Phys. Rev. B 26, 4449 (1982).
[16] S. Chaudhuri and K. K. Bajaj (unpublished).