Raman Scattering Resonant with Quasi-Two-Dimensional Excitons in Semiconductor Quantum Wells

J. E. Zucker[(a)]
Columbia University, New York, New York 10027

and

A. Pinczuk and D. S. Chemla
Bell Laboratories, Holmdel, New Jersey 07733

and

A. Gossard and W. Wiegmann
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 11 July 1983)

Raman scattering resonant with quasi-two-dimensional excitons in GaAs-(Al_xGa_{1-x})As heterostructures shows enhancement profiles in agreement with exciton structure measured in other optical spectra. The profiles have characteristic asymmetries in the incoming and outgoing resonances that are specific to the exciton–optical-phonon interaction in quantum wells. Direct evidence of the degree of exciton confinement appears in the Raman spectra.

PACS numbers: 71.38.+i, 71.35.+z, 73.40.Lq, 78.30.Gt

Excitons in multiple-quantum-well (MQW) GaAs-(AlGa)As heterostructures are formed by electron-hole pairs which are, to a variable degree, confined in the GaAs layers.[1] This reduction in dimensionality results in a pronounced enhancement of exciton behavior. Measurements of the optical properties of MQW's[1-3] have shown several sharp transitions which are characterized as excitons constructed from particle-in-a-well eigenstates. A number of recent papers have focused on various aspects of the physics as revealed by such quasi-two-dimensional excitations.[4-8]

In this paper we report the observation of Raman scattering resonant with excitons in GaAs-(AlGa)As MQW heterostructures. Although several previous Raman studies of GaAs-(AlGa)As heterostructures have been reported,[9-12] the work presented here is the first in which exciton behavior is observed. We find resonant Raman scattering (RRS) to be extremely effective as a probe of exciton properties in such systems of reduced dimensionality. The unique aspects of the quasi-two-dimensional exciton–optical-phonon interaction are revealed. New information on the degree of exciton confinement within the quantum wells is also obtained.

The selection of material parameters has an important effect on the quantum well potential and the degree of confinement of the exciton. The potential well depth is determined by the energy difference between GaAs and (Al_xGa_{1-x})As band gaps. For GaAs well thicknesses in excess of the bulk exciton diameter (~300 Å), the exciton is essentially three dimensional. On the other hand, if the GaAs thickness is less than ~50 Å inhomogeneous broadening due to imperfection of the interface smears the exciton feature.[3] A GaAs well thickness of ~100 Å insures both lower dimensionality and a sharp exciton. The width of the (AlGa)As barrier determines the coupling between electron and hole states in adjacent wells. When the barriers are wider than ~100 Å, coupling between the low-energy states of different wells is negligible for $x \gtrsim 0.15$. Thus excitons formed from these states are quasi-two-dimensional. However, for the highest levels, penetration of the wave function into the (AlGa)As layers can result in coupling between wells. Conduction- and valence-band states have been calculated in both the single-well and the Kronig-Penney coupled-well models. For our sample parameters the lowest-energy levels are essentially independent of which model is used since overlap between adjacent wells is minimal. Levels are labeled by the confinement quantum number n, with the lowest state being $n=1$. The exciton energies are estimated by means of the calculated quantum-well energy levels and the binding energies obtained by Miller *et al.*[4]

Two MQW samples grown by molecular-beam epitaxy were investigated. Sample A consists of 65 periods of 96-Å-GaAs–98-Å-($Al_{0.28}Ga_{0.72}$)As. Sample B has 77 periods of 102-Å-GaAs–207-Å-($Al_{0.27}Ga_{0.73}$)As. All measurements were made with the sample immersed in liquid helium at $T \lesssim 2$ K. Tunable cw dye lasers pumped by a Kr-

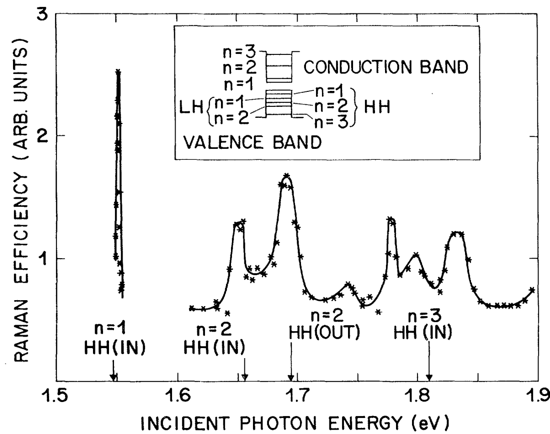

FIG. 1. Resonant Raman profile for a 102-Å MQW heterostructure (sample B). The arrows indicate exciton energies calculated with the single-quantum-well model.

ion laser served as the source of excitation. LD-700 dye was used in the range 685–885 nm[13] and Rhodamine-101 for 620–690 nm. The laser light at frequency ω_L was focused by a cylindrical lens and polarized along (110). It entered the sample along the (001) direction normal to the layers. Backscattered Stokes light at frequency ω_s and with polarization parallel to that of the incident beam was selected for this study. In this configuration the Raman spectra are dominated by forbidden scattering from LO phonons. This scattering is associated with Fröhlich exciton-optical-phonon coupling.[14,15]

Figure 1 shows the resonant Raman profile of sample B between 1.5 and 1.9 eV. Calculated energies of the $n=1$, 2, and 3 heavy-hole excitons are shown by arrows. The Raman spectra obtained in resonance with the $n=1$ and $n=2$ excitons show only the LO phonon of GaAs. This indicates that the envelope functions for these excitons are mostly confined to the GaAs layers. For incident photon energies near the $n=3$ and higher excitons we also observe the LO phonons of (AlGa)As, which is evidence that there is appreciable penetration of the barriers by these excitons.

We have compared the resonant Raman profiles with excitation luminescence spectra. This is shown in Fig. 2 for the $n=1$ excitons of sample A. In the energy range of the $n=1$ heavy-hole exci-

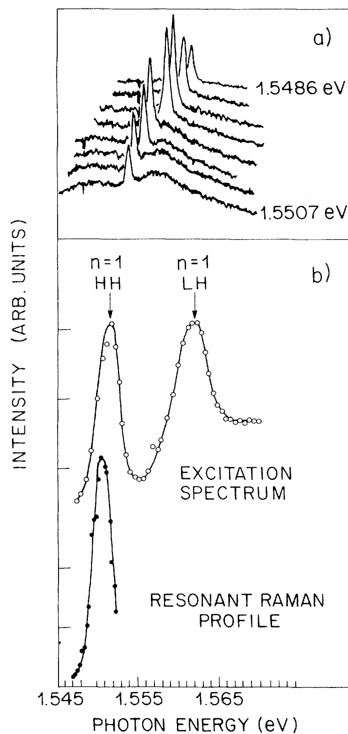

FIG. 2. (a) Resonant enhancement of the Raman spectrum from a 96-Å MQW heterostructure (sample A) as the incident laser is tuned near the $n=1$ heavy-hole exciton. (b) Comparison of the resonant profile with the excitation spectrum.

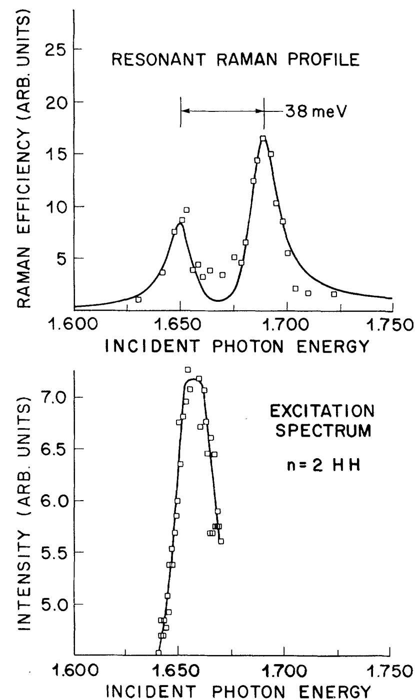

FIG. 3. Comparison of the resonant Raman profile with the excitation spectrum obtained from a 102-Å quantum-well sample in the energy range of the $n=2$ exciton. Data points are corrected for absorption. The solid line is a fit to the resonant profile with Eq. (2).

ton peak there is a clear correspondence between the excitation and Raman spectra. The resonant enhancement shown in Fig. 2, in which the incident photon energy coincides with that of the exciton, is termed the *incoming resonance.* A similar resonant behavior is also expected at the *outgoing resonance,* where the scattered photon energy equals that of the optical transition. Strong luminescence prevents us from observing the outgoing resonance of the $n=1$ heavy-hole exciton. The same luminescence obscures any $n=1$ light-hole exciton resonances.

This difficulty is not encountered for the $n=2$ and higher resonances since the major source of recombination emission occurs at the $n=1$ heavy-hole exciton. Separate incoming and outgoing resonances for the $n=2$ heavy-hole exciton of sample B can be seen in the results shown in Fig. 3, where we also compare the resonant Raman profile with the excitation spectrum. The separation between the two peaks in the resonant profile is close to the GaAs LO phonon energy ($\hbar\omega_{LO} = 36.6$ meV) as is expected for a double resonance. In most Raman scattering experiments in semiconductors the incoming and outgoing resonances cannot be separately resolved.[16] Double resonances are expected only in the presence of sharp optical structure, and thus are a characteristic signature of exciton behavior.[17-20]

The results presented in Figs. 1–3 establish the applications of RRS to the exciton spectroscopy of MQW heterostructures. The positions of the incoming resonances correspond to the energies of the optical transitions, while the occurrence of double resonance indicates that these excitations are narrow compared to a phonon energy. It is also possible to monitor the confinement of the exciton by the presence of (AlGa)As phonons in the Raman spectra. In the following analysis, the shape of the resonant profile is used to extract information on the coupling of optical phonons with the quasi-two-dimensional excitons.

Most striking in the profile of Fig. 3 is the pronounced asymmetry between the strengths of the incoming and outgoing peaks. Theories of RRS for three-dimensional excitons predict an asymmetric double resonance only when there exists a large dispersion of the refractive index.[21-23] This is not the case for our MQW heterostructures where the variation of the refractive index near the exciton resonances is of the order of a few percent.[24] Thus it appears that there must be another explanation for the asymmetry of the double resonance.

Near the $n=2$ exciton, the dominant contribution to the Raman intensity can be written as

$$I \sim \left| \sum_j \frac{\langle 0|P|j\rangle \langle j|H_{\rm ep}|2\rangle \langle 2|P|0\rangle}{(E_2-\hbar\omega_L)(E_j-\hbar\omega_s)} + \frac{\langle 0|P|2\rangle \langle 2|H_{\rm ep}|j\rangle \langle j|P|0\rangle}{(E_2-\hbar\omega_s)(E_j-\hbar\omega_L)} \right|^2 \qquad (1)$$

where $H_{\rm ep}$ is the Fröhlich exciton-phonon interaction, $\langle P\rangle$ represent the matrix elements for the optical transitions, and E_j is the energy of the exciton state $|j\rangle$. Within the effective-mass approximation[1,25] the numerators of both terms in Eq. (1) are identical. Since the term with $E_j=E_2$ can give no asymmetry to the resonant profile, we focus here on states with $j\neq 2$. It is clear that for these states the outgoing resonance, which is exhibited by the second set of terms, is largest when $E_j > E_2$. Thus the observed asymmetry in the Raman profile suggests that in Eq. (1) the states with $E_j > E_2$ are dominant. In fact the resonant profile can be interpreted in terms of the simplified expression

$$I \sim \left| \frac{1}{(E_2-\hbar\omega_L)(E-\hbar\omega_S)} + \frac{1}{(E_2-\hbar\omega_S)(E-\hbar\omega_L)} \right|^2 \qquad (2)$$

with $E > E_2$ as an adjustable parameter. The best fit is shown as a solid line in Fig. 3. This fit is obtained for $E = 1.85$ eV, which is close to the value calculated for the $n=3$ heavy-hole exciton: $E_3 = 1.81 \pm 0.04$ eV.

The above analysis indicates that exciton–LO-phonon scattering processes in which the exciton makes a transition to a different quantum-well state are important. This behavior can be explained by the quasi-two-dimensional nature of quantum-well excitons. In the quasi-two-dimensional limit exciton motion in the layer plane, which includes the relative motion as well as the translation of the center of mass, is completely decoupled from the degrees of freedom perpendicular to the layer. In this limit Raman backscattering, in which the LO phonon dipole moment has no in-plane component, can only result in a change in the exciton motion perpendicular to the layers, i.e., a change in quantum-well confinement number n. By contrast, in RRS associated with three-dimensional excitons wave-vector transfer to the exciton center-of-mass motion or changes in hydrogenic quantum numbers can occur.

In conclusion, resonant Raman scattering yields new insights into the physics of excitons in semiconductor quantum wells. The specific nature of the quasi-two-dimensional exction–LO-phonon interaction is revealed by asymmetry in the resonant profile. The degree of exciton confinement is monitored by the presence of the (AlGa)As phonons in the Raman spectra. As a spectroscopic tool the method is particularly advantageous in the observation of higher-lying transitions, which are obscured by background in other optical techniques.

We thank G. D. Aumiller for expert technical assistance.

[(a)]Work performed at Bell Laboratories, Holmdel, N.J. 07733.

[1]R. Dingle, W. Wiegmann, and C. H. Henry, Phys. Rev. Lett. 33, 827 (1974); see also R. Dingle, in *Advances in Solid State Physics,* edited by H. J. Queisser (Pergamon/Vieweg, Braunschweig, 1975), Vol. 15, p. 21.

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

[3]C. Weisbuch, R. C. Miller, R. Dingle, and A. C. Gossard, Solid State Commun. 37, 219 (1981).

[4]R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B 24, 1134 (1981).

[5]E. E. Mendez, L. L. Chang, G. Landgren, R. Ludeke, L. Esaki, and F. H. Pollak, Phys. Rev. Lett. 46, 1230 (1981).

[6]J. Hegarty, M. D. Sturge, C. Weisbuch, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. 49, 930 (1982).

[7]D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. 41, 679 (1982).

[8]G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Phys. Rev. B 26, 1974 (1982).

[9]P. Manuel, G. A. Sai-Halasz, L. L. Chang, Chin-An Chang, and L. Esaki, Phys. Rev. Lett. 37, 1701 (1976).

[10]G. A. Sai-Halasz, A. Pinczuk, P. Y. Yu, and L. Esaki, Solid State Commun. 25, 381 (1978).

[11]R. Merlin, C. Colvard, M. V. Klein, H. Morkoc, A. Y. Cho, and A. C. Gossard, Appl. Phys. Lett. 36, 43 (1980).

[12]C. Colvard, R. Merlin, M. V. Klein, and A. C. Gossard, Phys. Rev. Lett. 45, 298 (1980).

[13]G. D. Aumiller, unpublished.

[14]E. Burstein and A. Pinczuk, in *The Physics of Optoelectronic Materials,* edited by W. A. Albers (Plenum, New York, 1971), p. 33.

[15]R. M. Martin, Phys. Rev. B 4, 3676 (1971).

[16]M. Cardona, in *Light Scattering in Solids II,* edited by M. Cardona and G. Güntherodt (Springer-Verlag, Heidelberg, 1982), p. 19.

[17]J. Reydellet and J. M. Besson, Solid State Commun. 17, 23 (1975).

[18]T. C. Chang, J. Camassel, Y. R. Shen, and J. P. Voitchovsky, Solid State Commun. 19, 157 (1976).

[19]P. Y. Yu, in *Proceedings of the Thirteenth International Conference on Physics of Semiconductors*, edited by F. G. Fumi (Tipografia Marves, Rome, 1976), p. 235.

[20]A. Compaan and R. M. Habiger, in *Physics of Semiconductors 1978,* edited by B. L. H. Wilson (The Institute of Physics, Bristol and London, 1979), p. 489.

[21]D. L. Mills and E. Burstein, Phys. Rev. 188, 1465 (1969).

[22]B. Bendow, J. L. Birman, A. K. Ganguly, T. C. Damen, R. C. C. Leite, and J. F. Scott, Opt. Commun. 1, 267 (1970).

[23]A. S. Barker and R. Loudon, Rev. Mod. Phys. 44, 18 (1972).

[24]D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. Wiegmann, Appl. Phys. Lett. 42, 925 (1983).

[25]Yu. A. Romanov and L, K. Orlov, Fiz. Tekh. Poluprov. 7, 253 (1973) [Sov. Phys. Semicond. 7, 182 (1973)].