

Phonon satellite strengths in the photoluminescence spectra of a type-II GaAs/Al_{0.3}Ga_{0.7}As superlattice at elevated pressures

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We present results from a photoluminescence study of a GaAs/Al_{0.3}Ga_{0.7}As superlattice between 0 and 37 kbar. At 29 kbar, a type-I to type-II transition is observed, and this transition pressure is in good agreement with the predictions of the Kronig-Penney model. In the type-II regime, we detect direct $X\text{-}\Gamma$ recombination and have observed three distinct phonon satellites from structures having alloy layers. By studying the laser-power dependence of the photoluminescence spectra, we demonstrate that the X_z and X_{xy} level separation is just 8 meV, again in close agreement with theoretical predictions. Consideration of the relative intensity of the satellites shows that the strongest satellite at high laser powers is associated with the X_{xy} states and overlaps three further satellites coupled to X_z . These spectra provide experimental evidence that the X_{xy} states can have a significant effect on the optical emission from GaAs/(AlGa)As superlattices in the type-II regime, even when the conduction-band edge has X_z character.

I. INTRODUCTION

Due to the magnitude of the band offset that exists at interfaces between GaAs and AlAs and the fact that AlAs has an indirect band gap, the possibility of creating a type-II superlattice arises, in which electrons and holes are separated in both real and reciprocal space. This occurs if the GaAs layers are less than 12 monolayers (ML, 1 ML = 2.83 Å) thick, such that quantum confinement effects push the zone center, Γ electron states in the GaAs layers to a higher energy than the zone edge, X electron states in the AlAs layers.¹ Such Γ - X crossovers can also be arranged in more general (AlGa)As-based material systems. For AlAs/Al_xGa_{1-x}As structures, an essentially arbitrary superlattice can be designed to be type II through suitable choice of the alloy composition.² For GaAs/Al_xGa_{1-x}As systems, however, Γ - X investigations are limited, at ambient pressure, to alloy layers which are themselves indirect. This limits the fractional composition of the alloy to $x > 0.4$.

With the application of hydrostatic pressure, however, these limitations can be circumvented.³ Due to the different orbital character of the electronic states, the direct Γ - Γ band gap in (AlGa)As materials increases in energy with applied pressure, by around 11 meV/kbar, whereas the indirect X - Γ gap decreases at around 1 meV/kbar. Thus the properties of the X states in (AlGa)As-based superlattices become accessible even

when the sample is type I in the absence of pressure. The information that can be obtained from the many permutations of composition and pressure in this class of superlattice is important and complementary to the ambient pressure investigations which predominate.

The photoluminescence (PL) spectrum obtained from type-II crystals is strongly dependent on the nature of the lowest-energy X state.⁴ Due to the anisotropy present at the X point, a much higher effective mass exists in the longitudinal growth direction than in the transverse directions. Although quantum confinement effects would therefore dictate that the longitudinal states (X_z) would always be lower in energy than the degenerate transverse states (X_{xy}), these superlattices are almost exclusively grown on GaAs(001) substrates, which produces a biaxial strain in layers of (AlGa)As due to the finite lattice constant mismatch between these two materials. The effect of this strain is to reduce the energy separation of the transverse and longitudinal X states, and the magnitude of this reduction has been estimated at 23 meV for the case of binary AlAs layers.⁵ It is therefore possible to arrange for either X_z or X_{xy} states to be lower in energy, depending on the exact material composition and layer widths.

For the most widely studied GaAs/AlAs systems, the PL spectrum, from samples having X_z as their lowest states, shows a strong direct transition between electrons at X in the AlAs and holes at Γ in the GaAs. This process, which would usually be forbidden by momentum

considerations, is enabled by mixing of the Γ and X_z conduction-band wave functions. In addition to this strong transition, several weaker peaks are observed at lower energy, which correspond to momentum conserving processes involving the emission of certain phonons. In contrast, for structures having X_{xy} as their lowest state, it is these phonon satellites which dominate the PL spectrum, with virtually no direct X_{xy} - Γ recombination being observed.⁴

In this paper, we extend this analysis to account for the discrepancies which are observed between the PL spectra from GaAs/AlAs and GaAs/(AlGa)As structures having X_z as the lowest state. Published results on the former are numerous and indicate phonon satellites of around 1% of the direct transition intensity.^{6,7} The generality of this conclusion is well established in the work of Skolnick *et al.*,⁸ in which a saturation value of 1.5% was observed for a 12 ML/8 ML GaAs/AlAs sample at hydrostatic pressures well above the type-I–type-II transition. Though GaAs/(AlGa)As structures have not been so extensively studied, the phonon satellites are generally much more intense relative to the direct transition,^{3,9} rising to a saturation level of around 40% in the present work.

We have detected three phonon satellites, with the lowest-energy mode displaying a rapid increase in relative strength with increasing laser power. This we attribute to the near degeneracy of the X_z and X_{xy} states, which is predicted by calculations for our sample, but does not occur in thin-layer GaAs/AlAs samples due to the higher degree of quantum confinement. Such effects have been observed before, in the work of Feldmann *et al.*,¹⁰ in which the PL from a wider-layer GaAs/AlAs superlattice was observed to display very intense phonon satellites. This was attributed to the near degeneracy of the X_{xy} and X_z states on the basis of their calculated energies. Unlike our work, however, their sample had X_{xy} states which were at a lower energy than X_z and no conclusive experimental evidence was presented to support the calculations. In addition, the results presented by these authors indicated that only the X_{xy} states were involved with the phonon-assisted processes.

In this work, we present experimental evidence to show that, even when at a slightly higher energy than the X_z states, the X_{xy} states can significantly affect the PL spectrum obtained from GaAs/(AlGa)As superlattices. This conclusion is achieved by studying the laser power dependence of the PL spectrum between 0 and 37 kbar. We demonstrate that at high laser powers, the anomalously intense phonon satellite is associated with the X_{xy} states. We also demonstrate that the other two satellites appear to be associated with the X_z states, displaying a much weaker dependence on the incident laser power. It is presently unclear why the intensity of these X_z related modes should be so strong.

II. EXPERIMENT

The structure studied was grown by metal-organic chemical-vapor deposition on a semi-insulating GaAs substrate aligned along the (001) direction. 500 Å of

AlAs was grown as a buffer layer, onto which was deposited 50 repeats of a nominally 65 Å/60 Å, GaAs/Al_{0.3}Ga_{0.7}As intrinsic structure. The material was characterized by ambient pressure photoluminescence and Raman-scattering experiments. Although the folded acoustic modes detected in the Raman spectrum attested to the high material quality, a comparison of these results with the predictions of the Rytov model¹¹ indicated a slightly longer period than that nominally intended. By assuming that the alloy composition would be unlikely to vary significantly from its intended value, a careful fit to this model, coupled with the PL data, led us to conclude that the actual structure was closer to 75 Å/70 Å GaAs/Al_{0.3}Ga_{0.7}As.

The sample was thinned to approximately 40 μm by mechanical polishing with 6-μm grit paper and cleaved into sections measuring approximately 100 μm² for use with a diamond anvil cell.¹² The cell was pressurized using a helium gas bellows arrangement, permitting pressure variation at low temperatures.¹³ In practice, however, the cell was warmed to around 20 K, from its experimental temperature of 7 K, before changing the bellows pressure, in order to preserve the hydrostatic properties of the pressure transmitting medium. The pressure was calibrated using the PL from a small chip of ruby within the sample space, allowing pressure variation to within 1 kbar. No significant broadening of the ruby linewidth was observed, as would have been expected had the pressure application become nonhydrostatic.

The PL was excited using the 5145-Å line of an Ar⁺ laser, focused to a spot around 10 μm in diameter at the sample surface and detected in a backscattering configuration. The collected light was spatially filtered to limit the background signal often encountered with pressure cells and although the magnitude of this effect and of the accompanying reduction in the desired signal is difficult to quantify, we do believe that this approach leads to a significant improvement in the spectral resolution. The light was dispersed using a Spex 14018 double monochromator having 0.85-m stages, with the signal detected using standard photon-counting electronics. Optical alignment was performed with use of a charge coupled device imaging system, situated behind the entrance slit of the spectrometer, enabling the acquisition of very high-quality, reproducible, PL spectra.

III. RESULTS

The effect of hydrostatic pressure on the qualitative aspects of the PL spectra is shown in Fig. 1 for several selected pressures. In Fig. 2 we summarize all the collected data by plotting the energy of the detected peaks as a function of the applied pressure. The identification scheme for the peaks in this figure is given in the caption. As can be seen from Figs. 1 and 2, the sample is type I at pressures below 29 kbar, showing very intense recombination between electrons and holes at the Γ point in the GaAs layers (Γ_c^{GaAs} - Γ_v^{GaAs}). Several weaker peaks are detected in the spectra, originating from the bulk band gap of the GaAs buffer layer and from various acceptor levels.

In excess of 29 kbar the features of the spectra change dramatically, with the sample clearly exhibiting type-II behavior. Although recombination within the GaAs layers is still observed, a further direct process is detected between electrons at X in the (AlGa)As layer and holes at Γ in the GaAs layer ($X_c^{\text{AlGaAs}}-\Gamma_v^{\text{GaAs}}$). Accompanying this direct process is a series of phonon satellites at shifts from the direct transition of approximately 22, 32, and 46 meV. By assuming that the same phonon modes appear in our system as in the binary GaAs/AlAs case and following the recent resonant Raman measurements of Tribe *et al.*,^{14,15} we assign the 22- and 46-meV satellites to superlattice LA(X) and AlAs-like LO(X) phonons, respectively. In this notation, we use the term in parentheses to specify the point in the bulk Brillouin zone from which the phonon modes are thought to originate and retain the binary optical-phonon character because (AlGa)As is well established as exhibiting two-mode mixed crystal behavior.¹⁶ As pointed out in Ref. 14, however, the intermediate phonon assignment, at 32 meV in our system, is not so well established and may involve either confined GaAs-like optical modes or interface phonons. Nonetheless, we emphasize that to our knowledge this is the first observation of three distinct satellites in a

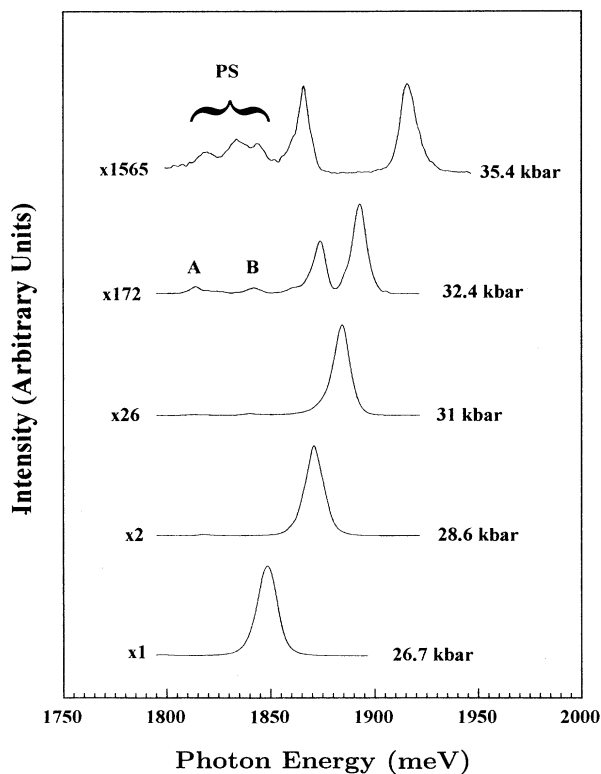


FIG. 1. Photoluminescence spectra as a function of pressure. The peaks marked B correspond to bulk levels, A to acceptors, and PS to phonon satellites. These assignments were made from analysis of the data displayed in Fig. 2, which established that the phonon satellites could not be attributed to shifts in acceptor or bulk levels.

type-II GaAs/(AlGa)As system at high pressures. Previous high-pressure investigations of structures with alloy layers have generally only resolved two satellites, at approximately 22 and 46 meV.⁹

The lines shown in Fig. 2 correspond to theoretical predictions of the type-I and type-II transition energies, as calculated using the standard Kronig-Penney model.¹⁷ This model is expected to be accurate for wide-layer samples, which have little or no coupling between adjacent wells; for our sample the lowest Γ miniband dispersion is predicted to be less than 0.5 meV. We assume a fractional valence band offset of $0.32\Delta E_g$ (where $\Delta E_g = [E_g^\Gamma]_{\text{AlGaAs}} - [E_g^\Gamma]_{\text{GaAs}}$),⁹ which does not vary with pressure, and use the parameter values given in Table I, with the alloy data being linearly interpolated from that of the bulk materials. Incorporation of the weak pressure dependence of the band offset reported by Lambkin *et al.*²² was found to have a negligible effect on our results. For the conduction-band X states and heavy-hole Γ states, it was assumed that the effective masses were independent of pressure. For the GaAs Γ conduction-band

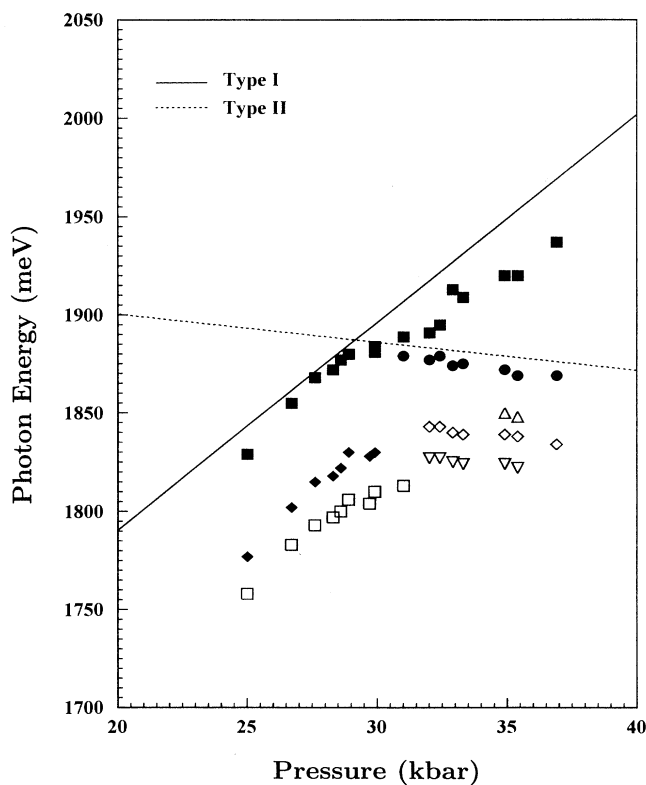


FIG. 2. Photoluminescence peak energies as a function of pressure. The solid line is the shift of the type-I energy gap and the dotted line that of the type-II gap, as predicted by the Kronig-Penney model. The peak assignments are as follows: \blacksquare , type-I direct Γ - Γ transitions; \bullet , type-II direct X - Γ transitions; \blacklozenge , bulk transitions from the GaAs buffer layer; \square , bulk acceptor levels; \triangle , type-II transitions involving superlattice LA(X) phonons; \diamond , type-II transitions involving phonons from the GaAs-like optical region; ∇ , type-II transitions involving AlAs-like LO(X) phonons.

TABLE I. Data used in the Kronig-Penney calculations. When not referenced, the data have been taken from Ref. 18.

Parameter	GaAs	AlAs
$E_g^{\Gamma-\Gamma}$ (0 kbar, 4.2 K)	1.519 eV ^a	3.13 eV ^a
$E_g^{\Gamma-X}$ (0 kbar, 4.2 K)	2.01 eV ^b	2.229 eV ^a
$dE_{\Gamma-\Gamma}/dP$	10.7 meV/kbar	10.7 meV/kbar ^c
$dE_{\Gamma-X}/dP$	-1.35 meV/kbar	-1.66 meV/kbar ^c
m_e^{Γ} (0 kbar)	0.067 m_0	0.15 m_0
$m_e^{X_z}$	1.3 m_0	1.1 m_0
$m_e^{X_{xy}}$	0.23 m_0	0.19 m_0
m_{hh}	0.62 m_0	0.76 m_0

^aReference 19.

^bReference 20.

^cReference 21.

states, however, a pressure-dependent mass was calculated following the method of Nunnenkamp *et al.*,²³

$$m_e^{\Gamma}(P) = m_e^{\Gamma}(0) \left[1 + \frac{\Delta_e^{\Gamma}(P) + [E_g^{\Gamma-\Gamma}(P) - E_g^{\Gamma-\Gamma}(0)]}{E_g^{\Gamma-\Gamma}(0)} \right] \quad (1)$$

in which the effective mass $m_e^{\Gamma}(P)$ and confinement energy $\Delta_e^{\Gamma}(P)$ were calculated self-consistently using the standard Kronig-Penney expressions. The confinement energies from these calculations, along with the pressure dependence of the bulk band gaps, yielded predictions of a type-I pressure dependence dE_I/dP of 10.57 meV/kbar, a type-II dependence dE_{II}/dP of -1.44 meV/kbar, and consequently a transition pressure P_t of 29.1 kbar. This is in close agreement with the experimental values, which indicate a transition pressure very close to this point. However, although a least-squares fit for the $X-\Gamma$ transitions produced a measured shift of -1.95 meV/kbar, in reasonable agreement with the prediction, a fit to the $\Gamma-\Gamma$ values was much less satisfactory. This is clearly due to the large sublinearity observed beyond the type-I-type-II transition, as can be seen in Fig. 2. We believe this is a genuine effect as opposed to an experimental artifact, due to the close agreement between theory and experiment for the $X-\Gamma$ transitions at these pressures and that the results presented in Fig. 2 were measured over a considerable time period and found to be easily repeatable. Considering just the low-pressure results, the $\Gamma-\Gamma$ peaks also display good agreement with theory, though it is hard to comment on the observed sublinearity beyond P_t .

A further prediction of the model is that, in the absence of strain, the X_{xy} states should lie 13 meV above the X_z states. Linearly interpolating the 23-meV downshift estimated for AlAs, for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers with $x=0.3$, leads to the prediction from the Kronig-Penney calculation of X_{xy} levels which are only 6 meV higher in energy than the X_z levels. This is an important result for understanding the effect of the incident laser power on

the PL spectra. The results of these calculations are summarized in Fig. 3, which shows the qualitative features of the predicted band alignment, at pressures beyond the type-I-type-II transition.

Results taken at 35.4 kbar, i.e., well beyond P_t , are shown in Fig. 4 for incident laser powers of 5 and 25 mW. Although with a laser spot around 10 μm in diameter this would appear to represent an extremely high power density, we point out that these are nominal laser powers, measured before reflection from an aluminum mirror and passage through several cryostat windows and a diamond. We would therefore expect a significant reduction in the actual laser power reaching the sample. Additionally, we can eliminate the possible influence of sample heating effects through temperature-dependent PL measurements. These indicated that at temperatures in excess of 10 K, the type-II direct transitions broadened dramatically, possible due to thermally activated delocalization of the spatially indirect excitons. As no similar

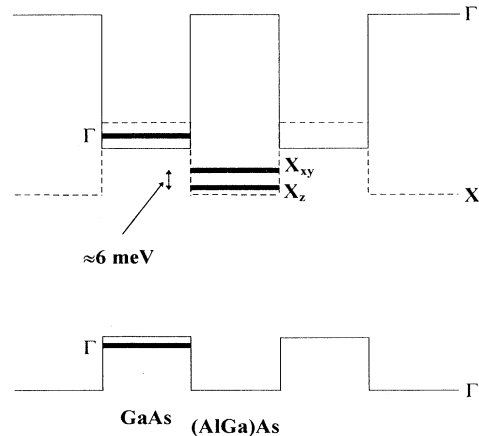


FIG. 3. The relative alignment of the confined electronic states at a pressure above the type-I-type-II crossover. Note the small energy difference between the X_z and X_{xy} states in the (AlGa)As layers (not to scale).

broadening was observed on increasing the laser power, we believe that the influence of sample heating can therefore be neglected. Considering first the 5-mW results, we detect the $\Gamma_c^{\text{GaAs}}-\Gamma_v^{\text{GaAs}}$ transition at 1920 meV, the $X_c^{\text{AlGaAs}}-\Gamma_v^{\text{GaAs}}$ direct transition at 1870 meV, and the three clearly resolved phonon satellites at the appropriate energy shifts from the $X-\Gamma$ transition energy. The ratio of the intensity of these peaks to that of the direct $X-\Gamma$ transition is around 40%, typical of these structures and much greater than in the GaAs/AlAs case.⁸

With higher excitation intensity, the change in the type-II spectrum is quite apparent. The line shape of the direct $X_c^{\text{AlGaAs}}-\Gamma_v^{\text{GaAs}}$ transition changes dramatically, with an asymmetric shoulder appearing towards higher energy. In addition to this, the appearance of the phonon satellites is altered, with the peak at 1848 meV enhanced to more than half the direct transition intensity, while the other two satellites are now visible only as shoulders on this dominant peak.

The explanation for this power dependence can be understood from the contrasting nature of the recombination from type-II GaAs/AlAs superlattices in which X_z or X_{xy} constitutes the ground state of the conduction band. In the former case, three phonon satellites are observed, having energy shifts of 28, 35, and 49 meV from the much stronger no-phonon line associated with the X_z state.^{7,8} The assignment of these modes has already been

discussed. In the latter case, the no-phonon transition from the X_{xy} states is observed only weakly because in a perfect crystal there is no mixing between these states and those at the Γ point in the GaAs layers. That this transition is observed at all is attributed to interface disorder.⁴ The PL spectrum is therefore dominated by the phonon satellites at energies of 12, 30, and 47 meV below the X_{xy} peak. The approximate intensity ratio of these satellites is, respectively, 1.0:0.7:1.4 and, again, following a recent work,¹⁴ we assign these peaks to superlattice TA(X), GaAs-like LO(X), and AlAs-like interface phonons, respectively.

If we make the assumption that the total scattering rate due to all phonon modes of a particular symmetry in an alloy is similar to the scattering rate from the identical mode in a binary, we can examine the relative intensities of the phonon satellites associated with the X_{xy} states in GaAs/(AlGa)As structures. While the TA(X) modes will be largely unaffected, due to the similarity of the acoustic bands in bulk GaAs and AlAs, the contribution from the AlAs interface modes and GaAs LO(X) modes will be substantially affected. In the former case, the magnitude of this change is very difficult to estimate. Although AlAs-like interface phonons are still observed in GaAs/(AlGa)As superlattices,²⁴ resonant Raman-scattering measurements have demonstrated that their coupling to the electronic system is a strong function of the interface quality.²⁴ In any case, the small aluminum concentration in the alloy will ensure that the contribution of these modes is relatively small. Regarding the GaAs LO(X) modes, we note that in the binary GaAs/AlAs case, this satellite is observed with comparable intensity to the other two phonon-assisted peaks despite the fact that the vibrational amplitude is mainly confined in the GaAs layers, while the electronic population is mainly located in the AlAs layers. Due to the two-mode mixed crystal behavior of (AlGa)As,¹⁶ however, vibrations of GaAs-like LO(X) character can be supported in the alloy layers of a GaAs/Al_{0.3}Ga_{0.7}As superlattice and so this mode ceases to be strongly confined. With a large vibrational amplitude present in the same layers as the X_{xy} electron population we may therefore expect a sizable increase in the magnitude of the GaAs-like LO(X) satellite. A further modification due to the alloy layers, which we believe may contribute to the phonon satellite structure, is the formation of a new interface mode branch of GaAs-like character not present in a GaAs/AlAs superlattice.²⁴ This branch is associated with a negative dielectric function in the alloy, similar to the branch of AlAs-like interface modes, so it is conceivable that these phonons may also couple to the electronic system in a similar way, producing a further spectral contribution at an energy around 3 meV higher than the GaAs-like LO(X) mode. Although it is difficult to confirm this contribution, as it would be unlikely that the spectral peak would be resolved from that of the LO(X) phonons, we believe that this may provide additional strength to the combined satellite. Finally, we still expect to observe a no-phonon transition and point out that the strict symmetry properties, which are the source of the momentum conservation rules which make it forbidden,

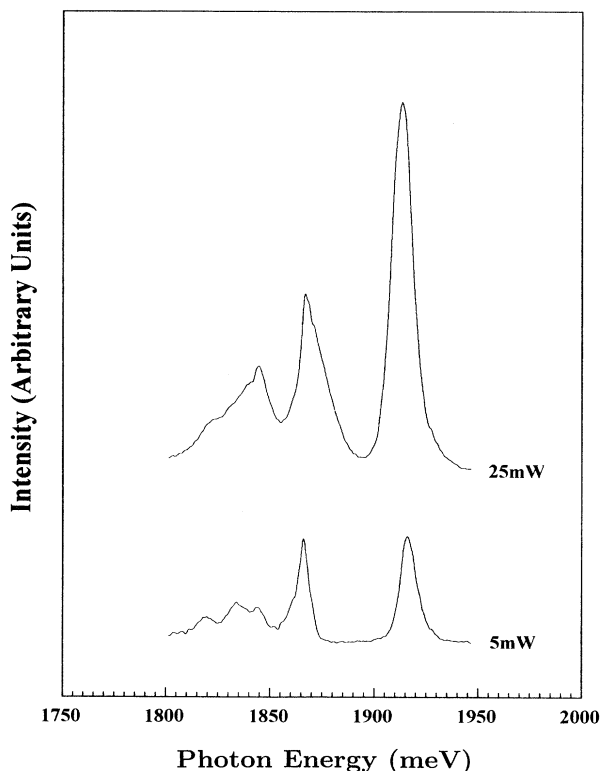


FIG. 4. Power dependence of the photoluminescence spectra at 35.4 kbar.

are partially removed by the natural chemical disorder of the alloy. This would enable direct transitions between X_{xy} and Γ , though the expected relative intensity of this peak is again difficult to estimate.

If we now consider the high-power result of Fig. 4, we note that the asymmetry observed in the X_z no-phonon line occurs at approximately 8 meV above the main peak. This is in good agreement with the theoretical prediction, from the Kronig-Penney model, that the X_z and X_{xy} states should be separated by 6 meV and is consistent with the discussion above, which suggests that X_{xy} - Γ no-phonon transitions should be observed. Further evidence that this is indeed the case arises from the discussion of the phonon satellite intensities associated with the X_{xy} states. The peak at 1848 meV, whose relative intensity increases significantly from low to high power, has a measured shift of approximately 30 meV from the X_{xy} transition, close to the energies of the GaAs LO(X) or interface phonons. [The exact phonon positions for GaAs/(AlGa)As structures at high pressure will be discussed later.] While we would also expect a strong satellite from the TA(X) modes, this would lie close to the X_z - Γ line and so would be difficult to resolve. The AlAs interface peak should be resolved, however, and, though of uncertain intensity, can be used to explain another significant difference between the low- and high-power results. In raising the power density there is a dramatic loss of resolution between the satellites at 1823 and 1838 meV, consistent with the predicted position of the AlAs interface phonon peak, which should lie between these two.

From this experimental evidence, we conclude that between 5 and 25 mW, the population of the X_{xy} states is greatly increased, probably due to the long radiative lifetime of carriers in the X_z states. As this effect becomes significant, the PL signal derived from the X_{xy} states is observed along with that from the X_z states, producing the power-dependent effects we have discussed. Therefore, due to their relative insensitivity to the incident laser power, we can establish that the modes at 1823 and 1838 meV are solely derived from the X_z state. We note that, although the satellite at 1848 meV displays a power dependence, this does not preclude the possibility that in the 5-mW spectrum this satellite is X_z related, with a quite separate satellite appearing at the same position, at high powers, but derived from the X_{xy} states. Indeed, we believe this is the case because there is no other evidence to suggest that there are any X_{xy} effects in the low-power result. Increasing the excitation power produces a combined contribution to the PL signal at this energy, creating the very intense satellite observed.

We emphasize that the preceding analysis is not based on the exact peak positions, but rather on approximate positions, coupled with an analysis of the relative intensities. This is an important point because, although we can make precise measurements of phonon energies in GaAs/AlAs structures at ambient pressure, the effects of both alloying and pressure will alter these energies. For example, for the GaAs LO(Γ) mode in the alloy, an upshift of 2 meV due to pressure¹⁹ is almost exactly bal-

anced by a similar sized downshift of 1.5 meV due to alloying.¹⁸ A similar pressure-induced upshift balanced by an alloying-induced downshift of ~ 3 meV (Ref. 24) is expected for the AlAs interface mode. However, the situation for the superlattice LA(X) modes is less clear, although it is again likely that the effects of pressure and alloying are approximately balanced. Nevertheless, since all the X_z satellite energies in the alloy system are observed to be between 4 and 6 meV lower compared with their values in GaAs/AlAs structures at ambient pressure, it would appear that some mechanism other than the alloying effect alone is responsible for this downshift. At present the origin of this mechanism is unclear. Notwithstanding this effect, we consider that allowing for a decrease in the phonon energies of a few meV, due to whatever cause, taken together with the relative intensities provides sufficient information to accurately identify the peaks.

Our results are a little surprising because the intensity of the X_z related satellites are approximately 40% of the no-phonon transition, in stark contrast to the 1–2% intensity of the same satellites observed from type-II GaAs/AlAs superlattices. In addition, it seems that this ratio has an unexpected dependence on the separation of the Γ and X_z states, rising to a maximum value at around 35.4 kbar before undergoing a marked decrease at higher pressures. Such behavior should be compared with measurements performed on GaAs/AlAs short-period superlattices by Skolnick *et al.*,⁸ who observed no similar effect. We are confident that this effect is genuine and that the spectral features do indeed arise from phonon-assisted recombination, as opposed to impurity related processes, because the evolution of the peaks with increasing laser power is quite unlike the expected saturation behavior usually associated with impurities. In addition, no hysteresis is observed as the pressure is cycled, as has been reported for pressure-activated deep levels in (AlGa)As-based materials.²⁵ We also believe that this unexpected ratio enhancement may be the key to explaining why this work represents the first identification of three phonon satellites in alloy systems because it is only very close to 35 kbar that sufficient resolution exists. A more detailed investigation of this resonant effect will be the subject of a future work.²⁶

IV. CONCLUSIONS

In summary, we have demonstrated that the intensity of the phonon satellites observed from type-II GaAs/(AlGa)As structures is very strong when compared with that observed from type-II GaAs/AlAs structures. From these measurements, we have observed three phonon satellites, derived from the X_z states, in such structures and obtained very strong evidence that the existence of nearly degenerate X_{xy} states can have a very significant effect on the optical emission, even when these

states lie at higher energies. This conclusion was reached by saturating the X_z states at high incident laser powers such that the effect of the X_{xy} states on the PL spectra could be identified. The observed power dependence of the various spectral features was used to identify their electronic origins.

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¹K. J. Moore, P. Dawson, and C. T. Foxon, *Phys. Rev. B* **38**, 3368 (1988).

²P. Dawson, B. A. Wilson, C. W. Tu, and R. C. Miller, *Appl. Phys. Lett.* **48**, 541 (1986).

³U. D. Venkateswaran, M. Chandrasekhar, H. R. Chandrasekhar, T. Wolfram, R. Fischer, W. T. Masselink, and H. Morkoç, *Phys. Rev. B* **31**, 4106 (1985).

⁴P. Dawson, C. T. Foxon, and H. W. van Kesteren, *Semicond. Sci. Technol.* **5**, 54 (1990).

⁵H. W. van Kesteren, E. C. Cosman, P. Dawson, K. J. Moore, and C. T. Foxon, *Phys. Rev. B* **39**, 13 426 (1989).

⁶E. Finkman, M. D. Sturge, and M. C. Tamargo, *Appl. Phys. Lett.* **49**, 1299 (1986).

⁷G. W. Smith, M. S. Skolnick, A. D. Pitt, I. L. Spain, C. R. Whitehouse, and D. C. Herbert, *J. Vac. Sci. Technol. B* **7**, 306 (1989).

⁸M. S. Skolnick, G. W. Smith, I. L. Spain, C. R. Whitehouse, D. C. Herbert, D. M. Whittaker, and L. J. Reed, *Phys. Rev. B* **39**, 11 191 (1989).

⁹D. J. Wolford, T. F. Kuech, J. A. Bradley, M. A. Gell, D. Ninno, and M. Jaros, *J. Vac. Sci. Technol. B* **4**, 1043 (1986).

¹⁰J. Feldmann, J. Nunnenkamp, G. Peter, E. Göbel, J. Kuhl, K. Ploog, P. Dawson, and C. T. Foxon, *Phys. Rev. B* **42**, 5809 (1990).

¹¹S. M. Rytov, *Akust. Zh.* **2**, 71 (1956) [*Sov. Phys. Acoust.* **2**, 67 (1956)].

¹²For a review of pressure cell techniques, see D. J. Dunstan and W. Scherrer, *Rev. Sci. Instrum.* **59**, 627 (1988).

¹³A. J. Shields, R. A. Woolley, P. C. Klipstein, J. Simmons, and R. H. Friend, *Semicond. Sci. Technol.* **4**, 301 (1989).

¹⁴W. R. Tribe, S. G. Lyapin, P. C. Klipstein, G. W. Smith, and R. Grey, *Superlatt. Microstruct.* **15**, 293 (1994).

¹⁵W. R. Tribe, Ph.D. thesis, Oxford University, 1994.

¹⁶I. F. Chang and S. S. Mitra, *Adv. Phys.* **20**, 359 (1971).

¹⁷H. S. Cho and P. R. Prucnal, *Phys. Rev. B* **36**, 3237 (1987).

¹⁸S. Adachi, *J. Appl. Phys.* **58**, R1 (1985).

¹⁹*Semiconductors. Physics of Group IV and III-V Compounds*, edited by K.-H. Hellwege and O. Madelung, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer-Verlag, Berlin, 1982).

²⁰D. J. Wolford and J. A. Bradley, *Solid State Commun.* **53**, 1069 (1985).

²¹K. Reimann, M. Holtz, K. Syassen, Y.-C. Lu, and E. Bauser, *Phys. Rev. B* **44**, 2985 (1991).

²²J. D. Lambkin, A. R. Adams, D. J. Dunstan, P. Dawson, and C. T. Foxon, *Phys. Rev. B* **39**, 5546 (1989).

²³J. Nunnenkamp, K. Reimann, J. Kuhl, and K. Ploog, *Phys. Rev. B* **44**, 8129 (1991).

²⁴A. K. Arora, A. K. Ramdas, M. R. Melloch, and N. Otsuka, *Phys. Rev. B* **36**, 1021 (1987).

²⁵W. P. Roach, M. Chandrasekhar, H. R. Chandrasekhar, and F. A. Chambers, *Phys. Rev. B* **43**, 12 126 (1991).

²⁶W. R. Tribe, P. C. Klipstein, R. A. Woolley, and J. S. Roberts (unpublished).