Uniaxial-stress investigation of the phonon-assisted recombination mechanisms associated with the X states in type-II GaAs/AlAs superlattices

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We present the results of a uniaxial-stress investigation of type-II GaAs/AlAs short period superlattices, using the optical techniques of photoluminescence (PL) spectroscopy and resonant Raman scattering. At ambient pressure, a resonance in the Raman cross section at the indirect, type-II band gap allows a direct measurement of the energies of the superlattice LA, GaAs-like LO, and AlAs-like LO phonons derived from the bulk X point. The positions of the Raman peaks are compared with the phonon-assisted PL transitions, indicating that the participation of GaAs-like LO phonons in the recombination process is not as well understood as has previously been thought. At sufficiently high in-plane stress, the transverse X minima become the conduction-band ground state, and the Raman measurements have again been used to examine the phonon modes, which can couple to these states. These measurements not only resolve an existing controversy in the literature, but they also demonstrate that one of the currently accepted assignments is incorrect. [S0163-1829(96)02231-X]

I. INTRODUCTION

Due to the band alignment in GaAs/AlAs superlattices, and the indirect nature of bulk AlAs, it is possible to use quantum confinement in short period structures to engineer a type-II system.¹ For AlAs layers of greater than 6 ML this occurs if the GaAs layer thickness is less than 12 ML.^{1,2} In such cases, the lowest Γ -electron level in the GaAs layers is at a higher energy than the lowest X level in the AlAs layers, producing electron and hole populations that are separated in both real and reciprocal space. In addition, the anisotropy of the X point in bulk AlAs produces different effective masses in the longitudinal and transverse directions, and therefore distinct electronic states in a superlattice structure. The properties of the longitudinal states, denoted X_{z} , and the transverse states, X_{xy} , are themselves quite different,^{3,4} so that the optical spectrum from a particular type-II sample is critically dependent on the nature of the lowest-lying X-electron level. It is the nature of the phonon interactions with the X-point electronic states, observed in the optical spectra, which is the subject of this paper.

Type-II behavior in $Al_xGa_{1-x}As/Al_yGa_{1-y}As$ structures has been established for several years,^{5,6} and the optical properties that are observed have formed the basis of many previous investigations.¹⁻¹³ While it was immediately appreciated that the heavy effective-mass X_z and light effectivemass X_{xy} states would be nondegenerate, due to the effects of quantum confinement,^{10,11,14} it was also quickly established that the influence of the biaxial compression in $Al_xGa_{1-x}As$ layers grown on GaAs substrates opposes the confinement separation.^{9,10,11} Consequently, in short period structures exhibiting large confinement effects, the X_z states are the ground state of the conduction band, whereas in wide layer structures, the strain effect has the greatest influence, and the X_{xy} states are at the lowest energy. The magnitude of the strain effect has been calculated, for the GaAs/AlAs system, to be 23 meV.³

Of particular relevance to the present paper is the work of Lefebvre *et al.*,¹⁰ who first demonstrated the effect of applying uniaxial stress in the plane of the layers of a type-II GaAs/AlAs short period structure. This approach can be viewed as modulating the internal compressive strain, so that the transverse *X* states in the direction of the applied external pressure reduce in energy very rapidly with respect to the longitudinal *X* states. Consequently, in a short period structure, a crossing can be induced between the longitudinal and transverse *X* states,^{10,15} permitting the properties of the latter to be studied in a much wider variety of samples. This approach is adopted in the present work to access separately the properties of both types of *X* state.

One of the main features of the optical spectra observed from both the longitudinal and transverse X states is the existence of a series of peaks to lower energies than the type-II band-edge excitonic transition. These have been noted in many previous studies,^{3,6,7,10–13} and have been assigned to phonon-assisted recombination, with general agreement that the form and relative intensity of the satellites is indicative of the nature of the conduction-band ground state.³ However, despite receiving attention in several publications,^{3,7,12,13} there remain a number of discrepancies in the precise peak assignments. Many of these difficulties stem from the fact that most of the observed phonons are thought to derive from the X point of the bulk Brillouin zone, coupling electronic states of bulk zone-edge character with virtual zone-center

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GaAs AlAs No. of GaAs buffer GaAs cap Growth Sample width (ML) width (ML) width (Å) width (Å) temperature (°C) repeats 1 11 8 100 2500 240 620 2 8 11 100 2500 250 615 3 10 10 10 2500 250 630 4 10 8 10 2500 250 610 5 8 8 100 250 2500 615

TABLE I. Summary of the notation and parameters of the samples.

states from which direct recombination can occur.¹² Consequently, the energies of the phonons, which are thought to participate, are only approximately known.

In this paper, we demonstrate how an interesting resonance in the Raman spectrum, which occurs at the X_{z} - Γ short band gap of type-II GaAs/AlAs period superlattices,^{16–18} can be used to provide a direct and highly accurate measurement of certain phonon energies derived from the X point of the bulk Brillouin zone. We then apply uniaxial pressure to change the character of the conductionband ground state from longitudinal to transverse X, thus providing complete consistency of the phonon modes between the two regimes. Comparison of the Raman measurements with the PL obtained below and above the transition pressure not only resolves the existing discrepancies in the literature, but also leads to a reassignment of one of the currently accepted phonon-assisted peaks.

II. EXPERIMENT

Experiments were performed on five superlattice samples grown by molecular-beam epitaxy on (001) GaAs substrates. 2500 Å of GaAs was deposited as a buffer layer, followed by the GaAs/AlAs superlattice, and a 240-250-Å GaAs capping layer. The growth temperature was between 610 and 630 °C, and the growth rates were calibrated using reflection highenergy electron diffraction. Raman measurements and PL measurements were performed on two of the samples, each having an 11-ML/8-ML GaAs/AlAs short period superlattice. Only one of these samples was subjected to external pressure. Two further samples were studied using photoluminescence (PL) spectroscopy as a function of uniaxial stress, and consisted of a 10-ML/10-ML and a 10-ML/8-ML superlattice, respectively. A further sample having a 8-ML/ 8-ML structure was examined only at ambient pressure using PL. The notation of these samples and their structures are summarized in Table I.

PL and Raman measurements were performed using a ring dye laser loaded with DCM, and optically pumped with 4 W all line output from an Ar⁺ laser. The error in calibration of the dye laser was estimated at ± 2 wavenumbers (cm⁻¹) [± 0.2 meV (Ref. 19)]. It was found experimentally that optimization of the Raman signal was very sensitive to the power density, and all Raman measurements were taken using a line focus, with around 50 mW of incident radiation. Brewster angle geometry was used, with the emitted light being analyzed by a Jobin Yvon T64000 0.65-m triple spectrometer, operating in multichannel mode.

In analyzing the PL spectra, the energies of the nophonon- and phonon-assisted recombination peaks were determined by careful fitting of a Gaussian line shape. This allowed the energy differences between no-phonon- and phonon-assisted peaks to be determined with an accuracy of $\pm 2 \text{ cm}^{-1}$. The precision in the phonon peak energies determined by the Raman measurements was obviously much better, and was limited by the 0.25-cm⁻¹ resolution of the spectrometer.

The application of uniaxial stress was achieved using a cell in which a force generated by a helium gas pressure is applied directly to the sample via a piston.²⁰ The pressure was determined through an accurate knowledge of the helium gas pressure, and the cross-sectional areas of the gas chamber and sample. The pressure cell was compatible with a standard Oxford Instruments liquid-helium cryostat, so that in all cases the temperature of the sample was 5 K.

III. RESULTS

In Fig. 1, we present the PL spectra obtained from sample 1 at pressures of 0 and 7.6 kbar, for which the conductionband ground state has, respectively, longitudinal and transverse X character. The pressure is applied along the (100)direction, splitting the degeneracy of the X_x and X_y states, as shown by the schematic evolution of all the important conduction-band states (relative to the top of the valence band), which is shown in the inset to the figure. We choose to apply stress along the (100) direction, because the inversion symmetry of a (001) oriented heterostructure is maintained, so that no piezoelectric fields are generated in the growth direction.^{21,22} Therefore, the electronic wave functions are not perturbed by changes in potential across a superlattice period, and the electron-phonon coupling can be directly compared with ambient pressure samples. The expected absence of piezoelectricity is confirmed by PL measurements performed up to 10 kbar, a detailed analysis of which has already been presented elsewhere.²³

The spectra of Fig. 1 are very similar to measurements performed on ambient pressure samples having the same relative band alignments.^{3,24} At 0 kbar, the very intense emission at 1790 meV is a direct recombination of electrons at X_z in the AlAs layers with holes at Γ in the GaAs layers, made allowed by the mixing of the Γ and X_z conductionband wave functions.²⁵ This peak remains visible at 1787 meV in the 7.6-kbar result, though its intensity is greatly reduced, due to the small occupation of X_z , which is no longer the ground state. At this pressure, direct recombination is also detected from the X_x state, and is seen as the comparatively weak emission at 1775 meV. Unlike the X_z -related direct recombination, this process is forbidden for the symmetry of a perfect superlattice, and is thought to be



FIG. 1. PL measurements of sample 1 taken at 0 and 7.6 kbar, below and above the crossover, respectively. The direct emissions are indicated by the nature of the X state from which they are derived, and the notation of the phonon satellites is established. The inset shows schematically the evolution of the four conduction-band states (Γ , X_x , X_y , and X_z), relative to the valence-band edge, under the application of uniaxial pressure along the (100) crystallographic axis.

activated by interface disorder.^{3,4,11} The peaks that are examined in this work lie at lower energies, and correspond to recombination processes involving phonon emission. We note that in both the X_z and X_x regimes three distinct phonon satellites are observed, and we adopt the notation established in Fig. 1. The energies of the phonons involved can be determined from the shifts of the satellites from the lowest direct recombination, and the conventional assignment of the participating phonon modes is presented in Table II.

Although, in principle, these assignments can be made easily through a direct comparison with established phonon energies, in practice, this approach is not so straightforward. Although recent low-temperature neutron-scattering experiments have determined the bulk GaAs dispersions with a high degree of accuracy,²⁶ they also demonstrate that the separation of the LO and LA branches at the X point is comparable to the linewidths of the phonon satellites in the PL. Consequently, whereas Dawson, Foxon, and van Kesteren³ assign σ_2 to an LA(X) vibration (where the term in parentheses indicates the wave vector of the bulk mode from which the superlattice phonon derives), Ge et al.¹⁵ conclude that it is related to the GaAs-like LO(X) mode. Although Zimmermann and Bimberg²⁷ concur with the former conclusion, they specifically point out that this assignment is uncertain. A further uncertainty encompasses the assignment of the γ_2 satellite. Smith *et al.*¹³ simply note that it is close to the GaAs LO branch, while only Skolnick et al.¹² consider the wave vector of the mode and conclude that it derives from a GaAs-like LO(X) phonon. Although there is currently no contention with the assignment of γ_3 and σ_3 , the AlAs phonon dispersion is not so well known, due to difficulties in preparing large volume samples for neutronscattering experiments.

The ideal approach to the assignment of these satellites is to measure the phonon energies of the superlattice using Raman scattering, and in order to access the vibrations which have the character of the bulk zone-edge X point, we use a recently discovered resonance, which occurs at the type-II X_z - Γ band gap of these superlattices.^{16–18} This resonance arises because of the allowed mixing between the X_z and Γ conduction-band wave functions, and produces strong scattering from the LA(X), GaAs-like LO(X) and AlAs-like LO(X) phonons, as well as from the more usually detected modes derived from the zone center. The Raman spectrum taken in the absence of pressure, and on resonance with the X_z -related band gap of sample 1, is shown in the upper curve of Fig. 2, along with the assignment of the important phonon peaks.

IV. PHONON SATELLITES COUPLED TO THE LONGITUDINAL X STATES

An analysis of the satellites coupled to the X_z states is relatively straightforward, because the type-II Raman resonance has the same electronic origins. In Fig. 2, we consider

TABLE II. Conventional assignments of the phonon satellites. The terms in parentheses indicate the bulk character of the superlattice phonon mode.

| Character of conduction-band ground state | Phonon satellite | Conventional assignment | Reference |
|-------------------------------------------------|------------------|-------------------------|-----------|
| $\overline{X_z}$ | γ_1 | LA(X) | 12,13 |
| X _z | γ_2 | GaAs LO(X)/ GaAs LO | 12/13 |
| X_z | γ_3 | AlAs $LO(X)$ | 3,12,13 |
| X_{xy} | σ_1 | TA(X) | 3 |
| X_{xy} | σ_2 | LA(X)/GaAs LO(X) | 3/15 |
| X_{xy} | σ_3 | AlAs $LO(X)$ | 3,15 |



FIG. 2. Direct comparison of the PL and Raman measurements from sample 1. The Raman spectrum is taken with the dye laser tuned exactly to the peak of the direct PL emission, and the assignment of the important Raman peaks is indicated.

the assignments graphically, plotting in the lower curves the PL measurements, and in the upper curve the Raman spectrum, measured with the dye laser tuned exactly to the energy of the direct PL peak. In this case, spectral features in the PL and Raman, which are coupled to the same vibrational mode, will be degenerate in energy, and dotted lines are drawn through several of the Raman peaks as a visual guide for the comparison. The measured energy shifts of the phonon satellites and of the Raman peaks, for all the samples studied in this work, are shown in Table III.

The first point to note is the excellent agreement, which is obtained between the γ_1 and γ_3 satellites, and the Raman peaks indicated by the dotted lines. This is supported by the

consistent measurements of the satellite energies in Table III, when compared with the Raman data, and we therefore conclude that these phonon satellites are correctly assigned in the existing literature. Of more interest is the γ_2 satellite, which is slightly lower in energy than the dotted line, drawn through the GaAs-like LO(Γ) phonon. This small energy difference suggests that the phonon assisting the recombination does not have the character of the bulk zone center. This is not surprising, because a zone-center phonon could only scatter an electron to an intermediate Γ state if its initial, real state, was also from the zone center. Although, in the case of the X_z state, this is possible due to the lowered symmetry of the superlattice, the character of the conduction band remains predominately that of the bulk zone edge, as is indicated by the observed coupling to the zone edge γ_1 and γ_3 phonons. If strong coupling was to occur through the GaAs LO band, it would, therefore, be expected from the X point, as assigned by Skolnick *et al.*¹² However, from the Raman measurements, such a phonon satellite would be expected at an energy shift of 247 ± 2 cm⁻¹, i.e., about 40 cm⁻¹ less than the measured energy. Furthermore, we believe that coupling to phonons, the wave vectors of which derive from between the bulk Γ and X points, and having intermediate energies, would be unlikely due to their mixed-symmetry properties.

Another interesting observation in the region of the γ_2 satellite is that as the sample period is decreased, the resolution between γ_1 and γ_2 is progressively lost, with only a single broad peak being visible from sample 5. This can be seen in the PL spectra of Fig. 3, which compares the satellite structure from samples 1, 4, and 5. In the case of sample 5, there is certainly some evidence that the γ_1 mode can still be detected at around 225 cm^{-1} , as indicated by the arrow in Fig. 3, which may suggest that the emergence of a single broad peak is attributable to the increased coupling to an intermediate phonon mode. Although it is difficult to use peak fitting routines in such poorly resolved features to obtain accurate peak positions, we note that the peak energy of the broad feature is observed at 247 cm^{-1} (see Table III), which is in close proximity to the GaAs LO(X) energy. There is also some indirect evidence for a fourth satellite in the samples which display well-resolved γ_1 and γ_2 features. If only three satellites are detected, and we assume that each has the same linewidth as γ_3 , then, in the 0-kbar spectrum of Fig. 1, we would expect significantly better resolution between γ_1 and γ_2 than is actually observed. Furthermore, our attempts at peak fitting, while allowing variation in the linewidths of γ_1 and γ_2 have also failed to reproduce the ob-

TABLE III. Comparison of PL and Raman measurements for the X_z -related phonon satellites.

| PL measurements (wave numbers, cm^{-1}) | | | nts m ⁻¹) | Raman measurements (cm ⁻¹) | | | |
|--------------------------------------------|------------|------------|--------------------------|----------------------------------------|---------------|---------------|---------------|
| Sample | γ_1 | γ_2 | γ_3 | LA(X) | GaAs LO(X) | GaAs LO(Γ) | AlAs LO(X) |
| 1 | 228 | 288 | 399 | 225.3 | 246.6 | 293 | 397.8 |
| 2 | 225 | 284 | 394 | 226.1 | 247.6 | 294.5 | 398.5 |
| 3 | 223 | 282 | 393 | | | | |
| 4 | 226 | 284 | 390 | | | | |
| 5 | 24 | 47 | 396 | | | | |



FIG. 3. Comparison of the PL measurements from samples 1, 4, and 5. Note that the direct PL emissions appear on a greatly reduced vertical scale relative to the phonon satellites. The arrow is discussed in the text.

served poor resolution. While ultimately inconclusive, these observations may at least suggest that the phonon satellite structure in the vicinity of γ_1 and γ_2 is more complicated than has previously been supposed.

Returning to the precise assignment of γ_2 , we believe that a possible solution may lie in coupling to the GaAs-like interface phonons, which have energies between the bulk GaAs LO and TO zone-center modes. These modes arise due to the electrostatic boundary conditions at each interface,²⁸ and a schematic of the dispersion relations (ignoring their interactions with confined LO phonons,²⁹ for both the GaAsand AlAs-like interface (IF) modes, is displayed in Fig. 4. Considering only the GaAs region, for phonon scattering between electronic states such as Γ and X_{τ} , which have no in-plane wave vector (k_{xy}) , the interface modes form two distinct bands, each extending between $k_z = 0$ and π/d , where d is the superlattice period. The upper band has a potential, which is even with respect to the center of the GaAs layer (indicated by the + in Fig. 4), and the lower band a potential that is odd (indicated by the -). For nearly equal GaAs and AlAs layer widths, the gap between these bands will be small, and so, for $k_{xy} = 0$, the upper band will extend from some lower value in the vicinity of 282 cm⁻¹ [midway between the GaAs LO(Γ) and TO(Γ) energies²⁶], to the GaAs LO(Γ) point of 293 cm⁻¹.²⁶

This lower (higher) energy, around 282 cm⁻¹ (293 cm⁻¹) corresponds to the even parity IF phonon with $k_z=0$ ($k_z=\pi/d$), which will couple via the Fröhlich potential to



FIG. 4. Schematic diagram of the interface mode dispersions in both the GaAs- and AlAs-like optical-mode regions, as a function of both k_{xy} and k_z . This diagram is for the specific case in which the GaAs layers are wider than the AlAs layers (Ref. 28).

the lowest X_{z} state, which lies at the superlattice zone center (boundary) in a perfect superlattice the period of which is an even (odd) number of monolayers. Since the dispersion of the X_{z} miniband is very small due to its very high effective mass, other X_{τ} states in the miniband with wave vectors up to the superlattice zone edge (center) may also be populated, and these will couple to the even parity IF phonons with k_{z} less than π/d and energies between 282 and 293 cm⁻¹. Thus, the strongest (weakest) contribution to the phonon satellite should be at the lowest energy with contributions decreasing (increasing) in strength up to 293 cm⁻¹. In a real superlattice, steps at the interface will ensure that different regions of the sample will exhibit even or odd character, so the observed satellite peak should lie somewhere between these two energy limits. This is in reasonable agreement with the phonon energies quoted in Table III, which lie between 282 and 288 cm⁻¹. Indeed, we note that the lowest value, of 282 cm^{-1} , occurs for sample 3, which has equal GaAs and AlAs layer widths, while the highest value, of 288 cm^{-1} , is from sample 1, which has the greatest difference in the layer widths. This is consistent with an assignment to IF phonons, because for equal layer widths there is no splitting between the two IF branches, giving a lower-energy limit of exactly 282 cm^{-1} , whereas the splitting progressively increases with the relative difference in the layer widths. If the IF modes do participate strongly in the phonon-assisted recombination, this raises the interesting possibility that coupling to the IF phonons may be enhanced, due to the recombination taking place across an interface. Regardless of this speculation, we shall present in the following section clear evidence that IF phonons can indeed assist the recombination process, supporting our contention that IF coupling to the X_{τ} states may also take place.

In concluding our analysis of the X_z satellites, we emphasize that the discussion of γ_2 is insufficient to make a positive assignment. Although we believe that the mode may be related to the GaAs-like IF branch, it is impossible to rule out coupling to either the GaAs-like LO(Γ) or LO(X) modes. Indeed, considering the loss of resolution between γ_1 and γ_2 in sample 5, we believe it is a strong possibility that coupling

TABLE IV. Comparison of PL and Raman measurements for the X_x -related phonon satellites.

| | PL measurements (cm ⁻¹) | | | Raman measurements (cm ⁻¹) | | | |
|--------|-------------------------------------|------------|------------|----------------------------------------|-------|---------------|---------------|
| Sample | σ_1 | σ_2 | σ_3 | γ_3 | LA(X) | GaAs LO(X) | AlAs LO(X) |
| 1 | 102 | 243 | 387 | 399 | 225 | 246.6 | 397.8 |
| 2 | | | | 394 | 226.1 | 247.6 | 398.5 |
| 3 | 93 | 241 | 377 | 393 | | | |
| 4 | 98 | 241 | 378 | 390 | | | |

to all three types of phonon is simultaneously observed, with the exact position and form of γ_2 dependent on the relative strengths of each. It may be difficult to answer this question experimentally, and there is a clear need for theoretical work to examine the relative strengths of the electron-phonon coupling for the different phonon modes, as the superlattice period decreases.

V. PHONON SATELLITES COUPLED TO THE TRANSVERSE X STATES

Unlike the satellites coupled to the X_z states, those coupled to X_x must be investigated indirectly, because there is no similar Raman resonance. However, the phonon energies are expected to undergo only small changes over the pressure range used in this work.³⁰ Resonant Raman measurements performed at 0 and 5 kbar confirm this expectation, with the change in the phonon energies being well within the ± 2 -cm⁻¹ error associated with the satellite energies to which they will be compared. We, therefore, assume that the change in the phonon energies is negligible over 7.6 kbar, and in Table IV make a direct comparison of the phonon energies measured by Raman scattering at 0 kbar, with the PL peak shifts at 7.6 kbar.

It is clear from the results of Table IV that the σ_2 satellite should be correctly assigned to a GaAs LO(X) vibration. Close agreement is observed between the phonon satellite and Raman energies in sample 1, and there is little variation of σ_2 in the three samples investigated. In addition, the energy of the LA band at the X point of GaAs/AlAs superlattices is expected to be almost independent of the sample parameters, because of the close similarity of the bulk acoustic bands. Therefore, the measured LA(X) energy is simply too low to account for the observed PL behavior, precluding its assignment to the σ_2 mode.

Another interesting feature of Table IV is a comparison of the energies of γ_3 and σ_3 . According to the conventional assignments given in Table I, these should derive from the same phonon mode. However, there is a consistent difference in the energy between γ_3 and σ_3 of at least 12 cm⁻¹, and although this is considerably less than the 50 cm⁻¹ linewidth of the spectral features, we point out that the shift is greater than the entire 10-cm⁻¹ dispersion of the bulk AlAs LO band,³¹ and that the σ_3 peak lies well below the established AlAs LO(X) mode. Therefore, since we have verified the assignment of γ_3 , it is apparent that σ_3 is also misassigned in the existing literature. Given that this satellite is not only at a lower energy than the entire bulk AlAs LO-phonon branch,

TABLE V. Proposed reassignment of the phonon satellites.

| Phonon satellite | Phonon assignment from present work |
|------------------|-------------------------------------|
| γ_1 | LA(X) |
| γ_2 | GaAs IF (?) |
| γ_3 | AlAs $LO(X)$ |
| σ_1 | TA(X) |
| σ_2 | GaAs $LO(X)$ |
| σ_3 | AlAs IF |

but is also significantly higher than the AlAs TO modes,³¹ we believe it may also be related to interface phonons. Certainly, it is difficult to recreate the energy of this peak by a combination of two phonons, and its strong intensity and narrow width would mitigate against multiple-phonon emission.

If the peak were related to interface phonons, we note that the necessary vibrational mode would have to couple an intermediate Γ state with the X_x states, which have a large in-plane wave vector, but $k_z = 0$. From the IF dispersions of Fig. 4, the requirement of large k_{xy} corresponds to the convergent energy of the different IF phonon branches, so the participating phonons should lie exactly midway between the zone-center AlAs LO and TO modes. This IF energy can be confirmed by resonant Raman measurements in the outgoing channel of the type-I band gap, because there is little overlap with the weakly dispersive AlAs LO modes, which may otherwise confuse the Raman spectrum.²⁹ Such measurements from sample 2 show a strong IF mode peak at approximately 381 cm⁻¹, as expected close to midway between the AlAs $LO(\Gamma)$ and $TO(\Gamma)$ energies. This value is in much better agreement with the σ_3 satellite measured from samples 1, 3, and 4, as can be seen from Table IV, strongly supporting the deduction that σ_3 is IF mode related. We note, however, that it is again impossible to rule out a contribution from the AlAs LO(X) phonon, which would enhance the intensity on the high-energy side of the σ_3 satellite.

The only mode which remains unexamined is σ_1 . Unfortunately, no Raman signals have been detected in the region around 100 cm⁻¹, and as such this is the only mode which we have been unable to consider experimentally. However, if we make the assumption that it is also derived from the *X* point, there would appear to be little alternative than a TA phonon, as previously reported.³ Certainly the energy of the mode is consistently between the GaAs and AlAs bulk TA(*X*) values [81 cm⁻¹ (Ref. 26) and ~110 cm^{-1,32} respectively], and would appear to be the least contentious of all the assignments.

VI. CONCLUSIONS

We have demonstrated that a number of the conventional assignments of the phonons, which assist type-II recombination processes, are presently incorrect, and we propose the assignments summarized in Table V. A number of these reassignments have been made on the basis of type-II resonant Raman measurements, which provide very accurate, direct values of the relevant X-point phonon energies. In this work, the role of uniaxial pressure has been crucial, not only in permitting the simultaneous study of the longitudinal and transverse X states, but also in confirming that the γ_3 and σ_3 satellites do not, as previously thought, derive from the same vibrational mode.

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- ¹⁹In this work, we retain the more standard energy units of meV when discussing PL peak positions, but adopt the units of cm⁻¹ when discussing phonon energies. This is to facilitate direct and accurate comparison with the technique of Raman scattering.
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