

Observation of quasidirect transitions in $\text{In}_{1-x}\text{Ga}_x\text{P}$ /graded GaP ($0.58 \leq x \leq 0.75$) alloys near the Γ - X_1 crossover

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We used room-temperature ellipsometry to study the quasidirect (no-phonon) transitions of *disordered* $\text{In}_{1-x}\text{Ga}_x\text{P}$ /graded GaP near crossover compositions ($0.64 \leq x \leq 0.75$) for the Γ_{1c} and X_{1c} conduction-band minima. Careful study of sample microstructures by transmission electron microscopy revealed no ordering. We identify the observed transitions at $(\Gamma_{15v}-X_{1c})$, $(\Gamma_{15v}-L_{1c})$, and $(\Gamma_{15v}-X_{3c})$. The enhanced amplitude of the quasidirect transitions is attributed to band mixing between Γ and L or X points in k space. This observation is corroborated by low-temperature photoluminescence and time-decay experiments. A two-level anticrossing behavior of the Γ_{1c} and X_{1c} gaps is observed in photoluminescence with Γ - X mixing energy $|V_{\Gamma-X}| \approx 12$ meV induced by random-alloy disorder.

I. INTRODUCTION

Measuring an indirect gap unambiguously is difficult because transitions are second-order processes involving phonons, thereby leading to overall transition-matrix element strengths some 3 orders of magnitude weaker than those found for direct transitions. In addition, it is difficult to detect indirect gaps because there are no singularities in the dielectric function, whereas direct gaps produce a singularity in the dielectric function.¹ However, when lattice periodicity is broken, indirect transitions can be observed as quasidirect or so-called no-phonon (NP) transitions.^{2,3} Translational symmetry can be broken in many ways, e.g., by defects, impurities,^{4,5} inherent potential fluctuation in alloys,⁶⁻¹⁰ chem-

ical modulation of alloys^{11,12} (e.g., alloy clustering, spontaneous ordering), the electron-hole interaction,¹³ and so on. For example, nitrogen isoelectronic dopants in $\text{Ga}_{1-x}\text{As}_x\text{P}$ and GaP form strongly localized deep levels associated with the X -point conduction-band minima.^{2,14} Quasidirect or NP transitions have been observed using photoluminescence (PL) for various semiconductor alloys, e.g., $\text{GaAs}_{1-x}\text{P}_x$,⁹ $\text{Al}_x\text{Ga}_{1-x}\text{As}$,^{8,10} and $\text{Si}_{1-x}\text{Ge}_x$ (Refs. 15 and 16) and were attributed to localized excitons trapped by compositional fluctuation arising from random-alloy disorder.

Here we report the observation of structures in room-temperature spectroscopic ellipsometric (SE) data from $\text{In}_{1-x}\text{Ga}_x\text{P}$ samples grown on compositionally graded layers on GaP that are attributed to quasidirect transi-

tions within the material near compositions where the L and X band cross Γ . We relate these structures to quasidirect transitions involving Γ_{15v} and X_{1c} , X_{3c} , and L_{1c} . This allows us to locate accurately these indirect minima relative to the top of the valence band. As far as we know, the positions of L_{1c} and X_{3c} of $\text{In}_{1-x}\text{Ga}_x\text{P}$ have not previously been detected unambiguously by any experimental methods, especially near room temperature. We suggest that these transitions arise from band mixing between Γ , and the L and X conduction-band minima due to random-alloy disorder, i.e., intrinsic alloy potential fluctuations. Our ellipsometry and photoluminescence measurements combined with careful characterization provides strong evidence that random-alloy disorder alone is the cause of the quasidirect transitions in $\text{In}_{1-x}\text{Ga}_x\text{P}$ alloys. Extrapolating our data to $x = 0.5$, we reassign the so-called “twin peaks” that were formerly associated with L (X) zone folding caused by CuPt-type ordering ($Y2$ structure) to the final states X_{1c} and the X_{3c} or L_{1c} .^{12,17}

We ascertained crossover explicitly with PL and its time-decay experiments at 1.4 K. The band mixing results in classic anticrossing behavior near crossover.^{17,18} For the Γ_{1c} and X_{1c} case, we fit the PL-determined energies and obtained a Γ - X mixing energy of $|V_{\Gamma X}| \approx 12$ meV. This anticrossing gap might have been formerly interpreted as an Γ - L indirect gap by Merle *et al.* using data inconsistent with ours.¹⁹

The results have direct implications regarding the crossover between direct and indirect gaps of $\text{In}_{1-x}\text{Ga}_x\text{P}$, which has been discussed by Bugajsky, Kontkiewicz, and Mariette.²⁰ Reported crossover compositions x_c vary from 0.64 to 0.75. Furthermore, there is no consensus about whether the lowest conduction bands of $\text{In}_{1-x}\text{Ga}_x\text{P}$ involve only the two conduction bands^{19–21} Γ_{1c} and X_{1c} or also L_{1c} . Our SE and PL results unambiguously support the Γ_{1c} , X_{1c} interpretation. Recent technological and scientific advances involving the $\text{In}_{0.5}\text{Ga}_{0.5}\text{P}/\text{GaAs}$ and $\text{In}_{0.5}(\text{Ga}_{1-x}\text{Al}_x)_{0.5}\text{P}/\text{GaAs}$ lattice-matched system make a knowledge of crossover composition x_c and the position of the lower conduction bands essential to further progress. In addition, there have been several band-structure calculations using the coherent potential approximation for the lower conduction bands for $\text{In}_{1-x}\text{Ga}_x\text{P}$ alloys for all compositions.^{20,22–24} Our identification of L_{1c} and X_{3c} as well as X_{1c} conduction bands should provide a good reference for further band-structure calculation of $\text{In}_{1-x}\text{Ga}_x\text{P}$ alloys.

II. SAMPLES AND EXPERIMENTS

The $\text{In}_x\text{Ga}_{1-x}\text{P}$ ($0.64 \leq x \leq 0.75$) samples were grown by hydride-vapor-phase epitaxy using HCl.²⁵ Details about sample growth and methods of characterizing composition are reported elsewhere.²⁶ We (three different groups) looked for evidence of long-range CuPt-type ordering and short-range ordering using transmission electron microscopy and surprisingly found none, although we observed composition modulation with an average period of 80–200 nm. Secondary-ion mass spectrometry with GaP as a reference showed no trace of impurities except Te ($n = 1 \times 10^{16}$ to 1×10^{17} cm⁻³). For samples 3, 4, and 5 (see Table I), the portions studied by SE and PL were on different parts of the wafer, and we label them differently, e.g., 3 (SE) and 3a (PL). We note that the average composition varied a couple of percent over the entire 37-mm wafer. The full width at half maximum (FWHM) of the x-ray rocking curve using high-resolution double-crystal x ray diffraction was 0.10° for the sample with $x = 0.715$, which was better than the reported value 0.14° by chin *et al.*²⁷ for a similar structure of $\text{In}_{1-x}\text{Ga}_x\text{P}/\text{graded GaP}$ grown by gas-source molecular-beam epitaxy. The lattice-matched $\text{In}_{0.5}\text{Ga}_{0.5}\text{P}/\text{GaAs}$ typically shows FWHM of 0.01° . The order of magnitude larger value of FWHM of graded samples originates from lattice defects caused by severe lattice mismatch.

Ellipsometry measures the change of the polarization state of light reflected at nonnormal incidence from the sample. This change is in turn related to ρ , the ratio of complex reflectance coefficients r_p and r_s , parallel and perpendicular component to the plane of the angle of incidence, respectively.^{28,29} Assuming a two-phase (ambient-substrate) model, the pseudodielectric function $\langle \epsilon(\hbar\omega) \rangle = \langle \epsilon_1(\hbar\omega) \rangle + i \langle \epsilon_2(\hbar\omega) \rangle$ is obtained from ρ . In order to enhance the structure present in the $\langle \epsilon(\omega) \rangle$ spectra, we calculated numerically the second derivative, $d^2\langle \epsilon \rangle / d\omega^2$, and obtained critical-point parameters by fitting to the resulting peaks the standard critical-point line shapes for direct gaps,³⁰

$$d^2\langle \epsilon \rangle / d(\hbar\omega)^2 = -n(n-1)Ae^{i\phi}(\hbar\omega - E + i\Gamma)^{n-2}, \quad n \neq 0, \quad (1)$$

$$d^2\langle \epsilon \rangle / d(\hbar\omega)^2 = Ae^{i\phi}(\hbar\omega - E + i\Gamma)^{-2}, \quad n = 0.$$

The critical-point parameters are the amplitude A , band-gap energy E , broadening Γ , and excitonic phase angle ϕ . The exponent n is equal to $(D-2)/2$, where D

TABLE I. Compositions and 300-K photoluminescence peak positions of $\text{In}_{1-x}\text{Ga}_x\text{P}/\text{graded GaP}$ samples.

Sample	1	2	3	3a	4	4a	5	5a	6	7
Ga, x	0.740	0.715	0.693	0.685	0.687	0.687	0.678	0.683	0.643	0.58
PL (eV), 300 K	2.263	2.220	2.180	2.168	2.172	2.172	2.154	2.164	2.096	2.002

is the dimension of the critical point. The critical points are those of the joint electronic density of states.

Photoluminescence and PL time-decay measurements were performed at 1.4, 10K, and room temperature. These measurements were performed using blue excitation from a frequency-doubled Ti:Al₂O₃ (4186.5 Å) laser with a pulse width of 1 ps and a repetition rate variable up to 82 MHz, depending upon the PL decay time. At 1.4 K, the average laser power was 20 μW at 82 MHz and the diameter of the laser spot size was 100 μm. All PL time-decay measurements were performed using the time-correlated single-photon counting technique with a temporal resolution of 500 ps.

III. RESULTS AND DISCUSSION

Figure 1 presents a plot of the second derivatives of the pseudodielectric function of In_{1-x}Ga_xP alloys from 1.5 to 3.0 eV. We can see structure between 2.0 to 2.5 eV. Positions of the E_0 edges measured by room-temperature PL were designated by arrows. We observed several structures that we associate with E_0 , $E_0 + \Delta_0$, E_{X1} , E_{L1} , and E_{X3} transitions and impurity related peaks. The E_{X1} and E_0 transitions overlap for the intermediate Ga compositions. We find that the critical-point line shape with a $D = 2$ direct gap gives the best fit for the quasidirect transitions of E_{L1} and E_{X3} as well as for the direct gap E_0 at 300 K.

Figures 2(a) and 2(b) show the plots of the $d^2\langle\epsilon\rangle/d(\hbar\omega)^2$ for $x =$ (a) 0.687 and (b) 0.693 samples of In_{1-x}Ga_xP/graded Gap. In Fig. 3(b), the $E_0 + \Delta_0$ transition is not discernible, possibly because of noise. We note

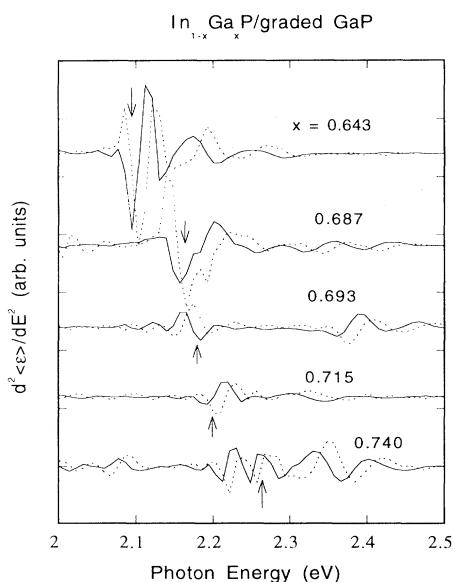


FIG. 1. Second-energy derivatives of the pseudodielectric functions for compositions $x = 0.643, 0.687, 0.693, 0.715,$ and 0.740 . Real and imaginary parts are shown as the solid and dashed lines, respectively. The arrows denote the E_0 gap measured by photoluminescence. We associate the thresholds with the E_0 , $E_0 + \Delta_0$, E_{X1} , E_{L1} , and E_{X3} , and the impurity-related peaks as described in the text.

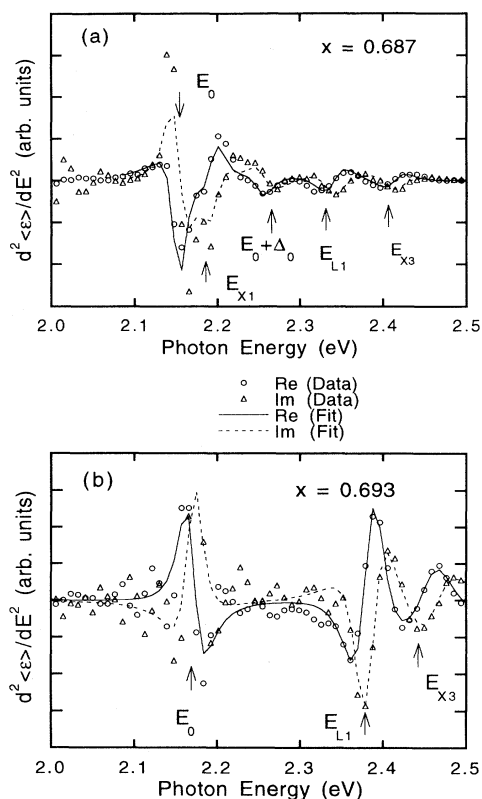


FIG. 2. $x = 0.687$ (a) and 0.693 (b) data of Fig. 1 shown as unconnected points along with best-fit line shapes.

that the direct and quasidirect transitions have approximately the same amplitudes. We carefully examined the strength of the quasidirect transitions with Te concentration and the period of composition modulation and found no correlation. Thus, these transitions do not appear to be due to the Te doping or the composition modulation.

Figure 3 presents a plot of the energy dependences of the E_0 , $E_0 + \Delta_0$, E_{X1} , E_{L1} , and E_{X3} transitions determined by the SE data. The error bars designate 95% reliabilities. For all samples except $x = 0.740$, PL measurements yield only the direct gap. For the $x = 0.740$ sample, room-temperature PL yields, in addition to a direct-gap peak, a weak no-phonon feature at 2.2 eV associated with X_1 . The room-temperature PL results are consistent with the SE data. Figure 3 shows that the crossover concentration at room temperature is $x = 0.724$ and is consistent with our RT PL. The fitted values for E_0 , $E_0 + \Delta_0$, E_{L1} , E_{X1} , and E_{X3} as a function of x are given by

$$\begin{aligned} E_0(x) &= 1.31 + 1.25x, \\ E_0 + \Delta_0(x) &= 1.54 + 1.04x, \\ E_{X1}(x) &= 1.84 + 0.52x, \\ E_{L1}(x) &= 1.80 + 0.78x, \\ E_{X3}(x) &= 1.85 + 0.83x. \end{aligned} \quad (2)$$

We identify the two peaks near 2.35 and 2.45 eV as E_{L1} , E_{X3} for the $x = 0.740$ sample. We exclude the possibility

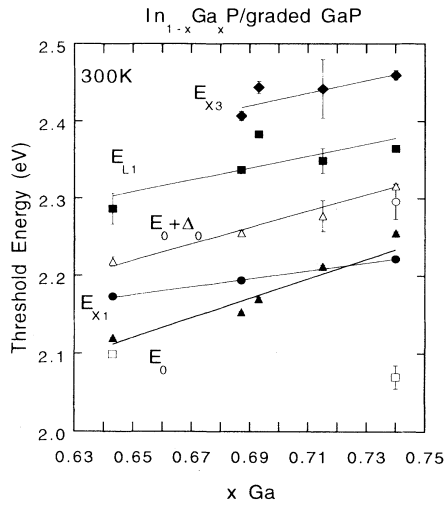


FIG. 3. Dependence of the energies of the E_0 , $E_0 + \Delta_0$, E_{L1} , E_{X1} , and E_{X3} thresholds as a function of Ga composition x . The impurity peaks associated with E_0 and $E_0 + \Delta_0$ are denoted by the unfilled circle and rectangle, respectively. The solid lines are weighted fits, except for E_{L1} .

of E_{X1} and E_{X3} because the binary end points InP and GaP show that the $E_{X3} - E_{X1}$ splittings are about 0.28 and 0.37 eV, respectively.³¹ The values for E_{X1} from the present work are 50 meV larger than those reported by Alibert *et al.*²¹ and Hakki, Jayaraman, and Kim.³² This is not surprising because $\text{In}_{1-x}\text{Ga}_x\text{P}$ band gaps are very much dependent on growth conditions because of order-

ing effects.¹¹

The extrapolated peak positions for E_{X1} , E_{L1} , and E_{X3} at $x = 0.515$ are 2.10, 2.20, and 2.27 eV, respectively. We note that these extrapolated gap positions match with the twin features reported for CuPt-ordered $\text{In}_{0.485}\text{Ga}_{0.515}\text{P}/\text{GaAs}$ by Kurtz¹⁷ using electroreflectance and photoreflectance. Our study suggests that the so-called twin features may arise partly from quasidirect transitions due to alloy disorder as well as the expected main cause, the zone-folding induced by CuPt ordering and $Y2$ structure.

In Fig. 1, the amplitudes of E_{L1} and E_{X3} increase and that of E_0 decreases as the Ga content increases to the crossover composition x_c of E_{X1} and E_0 . This suggests that strong level mixing between Γ_{1c} and $k \neq 0$ axis conduction band, e.g., X_{1c} , occurs because their energy levels are degenerate near the crossover region. Because the electronic eigenfunctions are formed by diagonalizing the Hamiltonian, and the wave-function mixing is more and more favored as the energy levels approach one another, even a small perturbation can lead to large mixing and consequently the transfer of oscillator strength from a strong level (E_0) to a weak level (E_{X1}).¹³ The level repulsion between E_0 and E_{X1} due to level mixing is not obvious in our RT ellipsometry data. The transfer of oscillator strength was indeed observed for the E_0 and E_{X3} crossover for $\text{GaAs}_{1-x}\text{P}_x$ alloys using the wavelength modulation reflectance technique.⁶ However, we note that E_{L1} and E_{X3} of $\text{In}_{1-x}\text{Ga}_x\text{P}$ alloys are relatively far away from E_0 by 0.1 and 0.3 eV, respectively.

Figures 4(a), 5(a), and 5(b) present PL and PL time de-

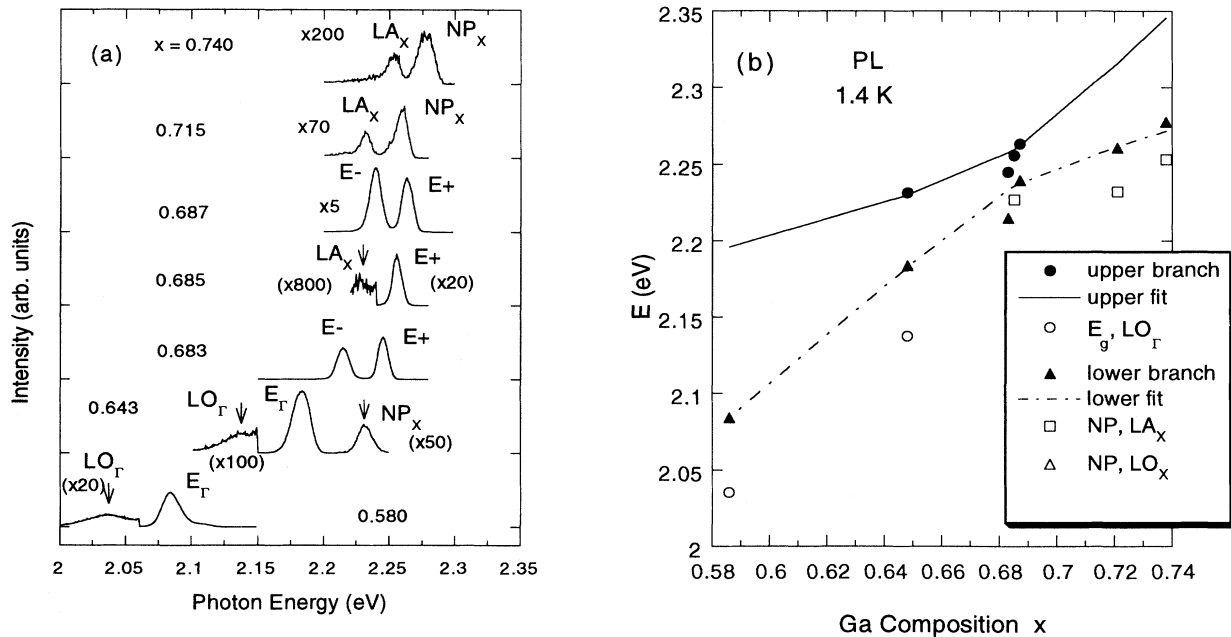


FIG. 4. (a) Photoluminescence data at 1.4 K. We denote the no-phonon exciton associated with the X_1 gap as NP_x , the direct-gap exciton at the Γ point as E_Γ , and their phonon sidebands as LA_x and LO_Γ , respectively. The Γ - X mixed states are denoted as $E+$ ($E-$) for upper (lower) branch. The peaks associated with no phonon and phonon sideband are magnified appropriately. (b) Dependence of the direct band gap, no-phonon peaks, and their phonon sidebands on Ga composition. The solid lines are the best fit of degenerate perturbation theory [Eq. (3)].

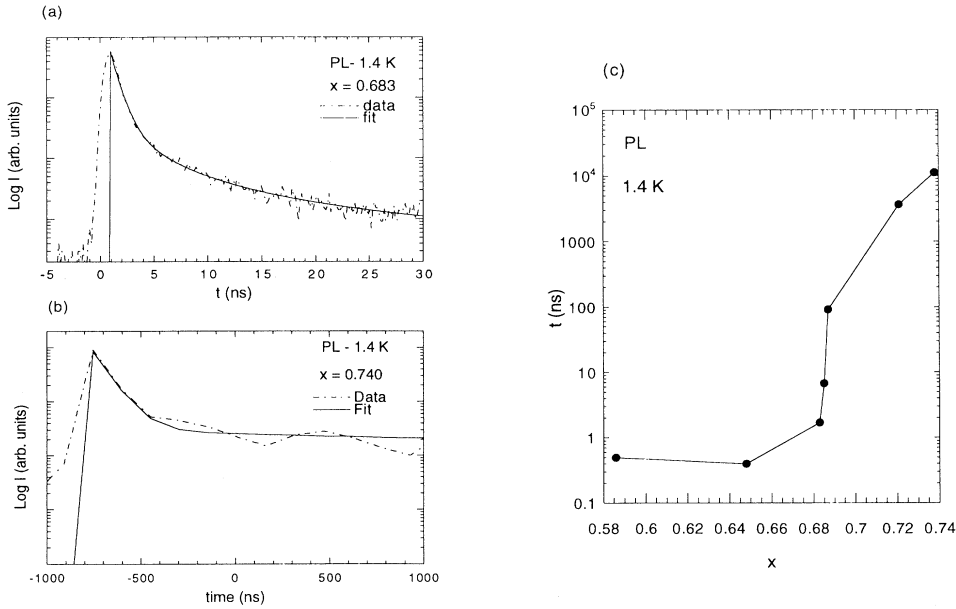


FIG. 5. Photoluminescence time-decay compositional dependence at 1.4 K for (a) intermediate regime ($x=0.683$), and (b) indirect gap ($x=0.740$) samples. Solid curves are calculated curves using the Klein-Sturge-Cohen theory of localized excitons by composition fluctuations and (c) is the plot of the fitted radiative lifetimes.

cays at 1.4 K. The crossover composition at 1.4 K is near $x=0.685$. The PL intensity and lifetime dictates three regimes: direct ($x < 0.65$), intermediate ($0.69 > x > 0.68$), and indirect ($x > 0.72$). The long-time tails were observed for indirect and intermediate regimes of Ga composition x , whereas long lifetime tails were absent for direct gaps. We fit all the time-decay data with the Klein-Sturge-Cohen (KSC) theory of localized excitons trapped by alloy composition fluctuations.³³ The KSC theory models the PL decay as having two components, radiative and nonradiative. Figures 5(a) and 5(b) show the plot of PL time-decay data. Calculated curves are shown as solid lines. The radiative decay time was at most 0.5 ns, as determined by our limited temporal resolution, for direct gap ($\Gamma_{15v}-\Gamma_{1c}$) materials and was as large as 11.2 μs for indirect gap ($\Gamma_{15v}-X_{1c}$) materials. The radiative decay time was between 1 ns and 1 μs for the intermediate regime.

In Fig. 4(b), the experimental values of the phonon sideband splittings match within ± 3 meV the calculated values of 44 meV (LO_X), 29 meV (LA_X), and 47 meV (LO_Γ) for $\text{In}_{0.7}\text{Ga}_{0.3}\text{P}$ using the linearly interpolated literature values for the binary endpoints.³¹ The nominally “forbidden” LO_Γ phonon sideband is allowed due to band mixing between Γ and X_{1c} . For the intermediate regime, the no-phonon peaks associated with X_{1c} (NP_X) and Γ (E_Γ) show similar time-decay behavior and have comparable amplitudes because Γ and X_{1c} are mixed by alloy disorder. The no-phonon peaks arise probably from excitons bound to Te donors. For example, the two peaks for $x=0.687$ are 23-meV apart, and the separation is too small to be explained as a phonon sideband.

To investigate the apparent mixing of the Γ and X_1 bands, we assume that near crossover these degenerate bands are coupled perturbatively. We use degenerate perturbation theory for two bands as adopted by Kelso *et al.*¹³ and fit the peak energy positions of the 1.4-K PL

data with the equation

$$E = \frac{E_{0\Gamma} + E_{0X}}{2} + \frac{1}{2} \sqrt{(E_{0\Gamma} - E_{0X})^2 + 4V_{\Gamma X}^2}. \quad (3)$$

Using the Onton and Chicotka 2-K PL expression³⁴ for $E_{\Gamma 1} = (1.409 + 0.659x + 0.758x^2)$ eV and our own offset RT SE result $E_{X1} = (1.890 + 0.52x)$ eV, we find that $|V_{\Gamma X}| \approx 12$ meV gives the best fit. A Γ - X mixing energy of $|V_{\Gamma X}| = 6$ meV was found by Capizzi *et al.*¹⁸ for bound excitons in $\text{GaAs}_{1-x}\text{P}_x$ alloys, although they did not observe the upper branch of the mixed state. The factor of 2 difference in the $|V_{\Gamma X}|$ between $\text{In}_{1-x}\text{Ga}_x\text{P}$ and $\text{GaAs}_{1-x}\text{P}_x$ is consistent with the factor of 2 difference in the electronegativity difference of In, Ga, and As, P in the two alloys.³⁵

Determination of a mechanism for coupling these degenerate bands is difficult. One obvious possibility is the short-range alloy ordering. Such a phenomenon may optically couple these bands but it may not show up in the electron microscopy images. We are currently studying a possible ordering by scanning tunneling microscopy. A second possibility is alloy disorder, which in principle mixes the two states. The strength of the mechanism may be estimated using second-order perturbation theory. We have calculated the absorption coefficient, which is closely related to the imaginary part of dielectric function, using the tight-binding theory³⁶ to estimate the enhancement of the strength of quasidirect transition near-band-gap crossover. There are three enhancement factors: (i) Δ^{-1} increases near-band-gap crossover ($\Delta = E_\Gamma - E_X$) due to transfer of oscillator strength from Γ to X_1 , (ii) a large density of states of X conduction-band minima, $\rho_X(E)$, and (iii) a large electronegativity difference for In and Ga [$V_{\text{In}} - V_{\text{Ga}} = 0.56$ eV compared with that of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ (0.18 eV) or $\text{GaAs}_{1-x}\text{P}_x$ (0.31 eV)].³⁵ However, the absorption strength is still 2 orders of magnitude smaller than that of the direct transition

when $\Delta=0.1$ eV. Furthermore, the perturbation theory does not predict the direct-gap-like singularity fitted by the critical-point line shape in the $d^2\langle\varepsilon\rangle/d\omega^2$ spectra. The $\langle\varepsilon\rangle$ is basically a measure of a joint density of states.¹ In terms of the second-order perturbation theory, the joint density of states for quasidirect transitions due to alloy disorder is proportional to $\sim(E-E_g)^n$ for $E > E_g$ and 0 for $E < E_g$ where $n=1$ ($2D$) and 2 ($3D$), and so has no discontinuity in the $\langle\varepsilon\rangle$ spectra, contrary to the step function densities of $D=2$ direct gap.

A third explanation involves alloy disorder, treated nonperturbatively, which creates resonant states. Random-alloy disorder causes mixing of the wave function of Γ with X or L . In principle, it can produce a bound level ($E_{\text{indirect}} < E_0$) or a resonance level ($E_{\text{indirect}} > E_0$) near the Γ point. Our data may suggest that the mixing of wave function between Γ and, for example, L_1 , due to the formation of resonance level can be substantial.³⁷ Furthermore, this resonant-level hypothesis gives direct-gap-like singularity in $d^2\langle\varepsilon\rangle/d\omega^2$ spectra by producing a step-function-like density of states like that of the $2D$ direct gap.

IV. CONCLUSION

We have observed the quasidirect (no-phonon) transitions of disordered $\text{In}_{1-x}\text{Ga}_x\text{P}$ /graded GaP ($0.64 \leq x \leq 0.75$) by spectroscopic ellipsometry at room temperature. We identify these transitions as $(\Gamma_{15v}-X_{1c})$, $(\Gamma_{15v}-L_{1c})$, and $(\Gamma_{15v}-X_{3c})$. The enhanced amplitude of the quasidirect transitions is attributed to band mixing between the Γ and the X and L points in k space. Our PL data confirms quasidirect transitions and we found anticrossing behavior between Γ_{1c} and X_{1c} with mixing energy $|V_{\Gamma X}| \approx 12$ meV.

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