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Effect of composition and pressure on the nitrogen isoelectronic trap in $GaAs_{1-x}P_x$ †

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The luminescence and absorption behavior of the nitrogen trap in $GaAs_{1-x}P_x$ is investigated as a function of composition and pressure. Luminescence and absorption data support the identification of strong phonon coupling to the broad N_X band (formerly identified as NN-pair recombination transition for $x \lesssim 0.85$). Pressure measurements in the composition range $0.38 \lesssim x \lesssim 0.48$ demonstrate, in a continuous manner, the behavior of the N_Γ transition (previously identified as NN_3 for crystal composition $x = 0.37$ and 0.38 and the A line for $0.40 \lesssim x \lesssim 0.55$). The oscillator strength of this state for an $x = 0.48$ $GaAs_{1-x}P_x$:N sample is observed to decrease with increase in pressure, disappearing at ~ 6.6 kbar or a simulated composition $x \sim 0.55$ ($E_\Gamma - E_N \sim 140$ meV).

INTRODUCTION

Recently, the $GaAs_{1-x}P_x$ alloy system doped with nitrogen has become important for practical[1-3] and fundamental reasons.[4-8] For both reasons it is important to establish the position (energy) of the nitrogen states as a function of alloy composition, or in other words with respect to the Γ and X band edges of the system. While the important effect of the N trap on the 300 °K electroluminescence efficiency is relatively well known,[1-3] recent studies[9] of $In_{1-y}Ga_yP$:N and more particularly N-implanted $GaAs_{1-x}P_x$ (Ref. 10) have shown that the luminescence transition originally identified with the A line ($x \approx 0.35$),[11] and later with the A line (Fig. 4 of Ref. 12) and NN pairs ($0.30 \sim x \lesssim 1$),[12-14] is in fact due to recombination at single N impurities (A line) perturbed by the alloy (As-P or In-Ga) disorder. Apart from alloy disorder, the broad nature of this transition (here labeled N_X) is attributed to a strong phonon sideband that is characteristic of an impurity strongly coupled to the lattice.[9,10] In this work we present further data illustrating the "A line" behavior of the broad N_X band in the indirect region ($x \gtrsim x_c$) of $GaAs_{1-x}P_x$:N.

Since the N_X band of the present work exhibits A-line properties,[10] the "A line" observed earlier at higher energy near the Γ band edge [$x = 0.35$ (Ref. 11); $0.40 \lesssim x < 0.55$ (Ref. 13)], and that ($E_N \lesssim E_\Gamma$) previously identified as NN_3 in $x = 0.37 - 0.38$ crystal,[15] are of different origin and character.[16] As the data below show, this level is a bound state associated with the Γ conduction band. Pressure measurements are described showing that this state (N_Γ) follows the Γ minimum for $x \lesssim 0.45$ and then beyond the direct-indirect transition ($x_c = 0.45, 77$ °K) bends and tends to parallel the X band edge until at $x \sim 0.55$ (where $E_\Gamma - E_N \sim 140$ meV) it disappears.

EXPERIMENTAL METHOD

As in previous work,[12] the $GaAs_{1-x}P_x$:N of the present work has been grown by the now standard AsH_3-PH_3 vapor-phase epitaxial (VPE) process.[17] Nitrogen is introduced into the crystals by adding NH_3 to the gas flow. Thin samples for absorption or photoluminescence experiments are polished and etched from the as-grown crystals. For the pressure measurements of this work N-doped $GaAs_{1-x}P_x$ substrates are prepared into $In_{1-y}Ga_y$-$P_{1-z}As_z$-$GaAs_{1-x}P_x$:N single heterojunctions[18] that do not absorb the higher-energy side of the electroluminescence spectrum where an "A line" might be observed.

A Cary Model No. 14RI spectrophotometer is used for absorption measurements on thin (~30-μm) homogeneous samples. For photoluminescence measurements, a Spectra Physics Model No. 165 Ar^+ laser is used as an excitation source; a Spex Model No. 1702 0.75-m grating monochromator and an RCA 8645 photomultiplier are used to examine the recombination radiation.

For the pressure measurements described below, a helium-gas pressure system[19] and a pressure vessel with a sapphire optical window[20] are used. The pressure vessel is submerged in a liquid-nitrogen bath. Fiber optics are used to collect the diode luminescence and to direct the light to a Jarrell-Ash 0.5-m grating monochromator fitted with an RCA C31034 photomultiplier. A manganin gauge is used for pressure measurements. The pressure coefficients of the Γ and X band gaps of $GaAs_{1-x}P_x$ are known from previous work[21] and from our own data obtained on N-free diodes, and are $dE_\Gamma/dp \approx +11\times10^{-6}$ eV/bar and $dE_X/dp \approx -1\times10^{-6}$ eV/bar. The combined pressure variation of the band gaps simulates a composition change of $dx/dp \sim 1.1\%$/kbar for $GaAs_{1-x}P_x$.

EXPERIMENTAL RESULTS

In the indirect region of $GaAs_{1-x}P_x$:N ($x>x_c \approx 0.45$, 77 °K) the N trap exhibits the luminescence spectra (77 °K) and A-line behavior shown in Fig. 1. As shown, all the curves are plotted as a function of energy with the high-energy luminescence peaks used as a common reference. The spectrum (a) of GaP:N ($x=1, n_N \sim 2\times10^{18}$ cm^{-3}) possesses the sharp A line, NN_3, NN_1 and associated phonon-replica transitions expected.[22] For the case of the $x=0.90$ $GaAs_{1-x}P_x$:N ($n_N \sim 10^{18}$ cm^{-3}) sample of Fig. 1(b) the phonon sideband that has recently been shown to be characteristic of the N trap in both $In_{1-x}Ga_xP$:N[9] and $GaAs_{1-x}P$:N[10] is clear. We note that in earlier work the LO-phonon replica of the N_X transition has been assigned to NN_3 pairs, and the N_X-2LO peak to NN_1.[12,13] At still lower composition ($x=0.84$) the luminescence spectrum, as shown in Fig. 2(c) ($n_N \sim 5\times10^{17}$ cm^{-3}), exhibits less structure in the phonon sideband owing to the larger exciton binding energy and the resulting increased electron-phonon interaction. In previous work ($x=0.84$) the assignment NN_3 for N_X has been made, and NN_1 for N_X-LO.[12,13]

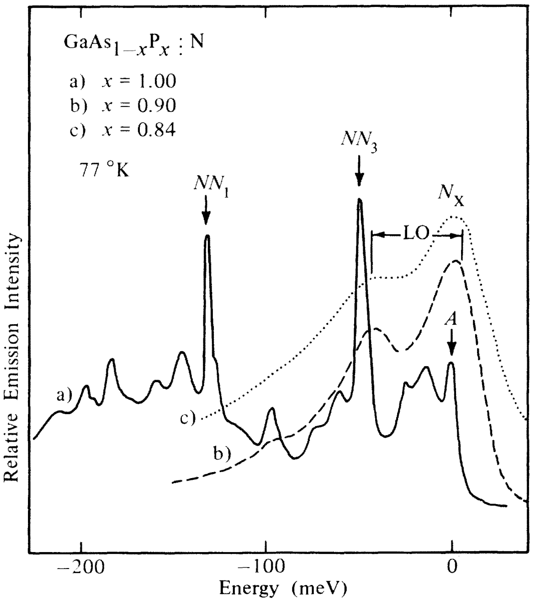

FIG. 1. Photoluminescence spectra (77 °K) of (a) GaP:N, (b) $x=0.90$ $GaAs_{1-x}P_x$:N, and (c) $x=0.84$ $GaAs_{1-x}P_x$:N. The high-energy side of the A line or N_x peak is taken as the reference for the spectra of the three crystals. Note that as the crystal composition x decreases, the spectra broaden owing to As-P disorder, increased electron binding energy, and stronger electron-phonon interaction.

Agreeing with the photoluminescence data of Fig. 1, Fig. 2 shows the expected sharp A-line absorption peak of GaP:N (a), and the broadened absorption (b) and luminescence (c) spectra of an $x=0.75$ $GaAs_{1-x}P_x$:N sample. The Stokes shift between the absorption and luminescence peaks, which is characteristic of an impurity level strongly coupled to the lattice,[23,24] has been previously demonstrated in[9] $In_{1-x}Ga_xP$:N and, although not recognized as such, in $GaAs_{1-x}P_x$:N.[12] All the spectra of Fig. 2 are plotted as a function of energy with the X band edge used as a reference.

Because the absorption and recombination at an isoelectronic center are dominated by exciton ef-

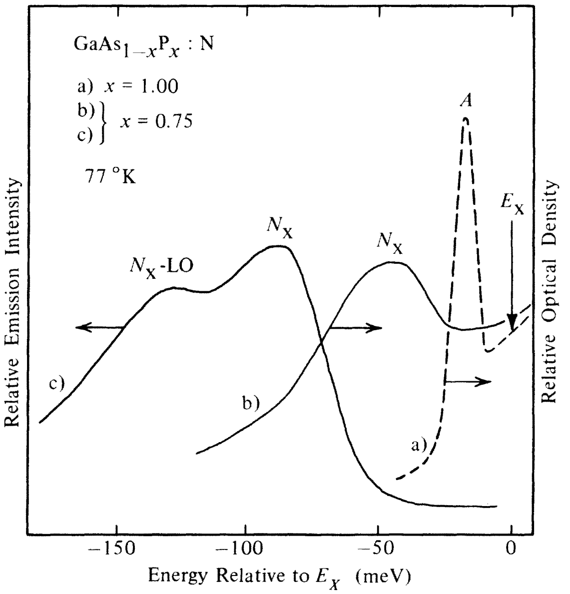

FIG. 2. Absorption spectra (77 °K) of (a) GaP:N and (b) $x=0.75$ $GaAs_{1-x}P_x$:N. Curve c shows the luminescence spectrum of the alloy. The broader absorption and emission spectra of the alloy, and the Stokes shift between them (b) and (c), are characteristic of an impurity strongly coupled to the lattice.

fects,[22] the initial and final states can be treated as discrete, and the conventional configuration-coordinate model can be applied to determine the spectral half-width.[23,24] This gives for the temperature variation of the N_x absorption-emission bandwidth the expression

$$W_{1/2}=\alpha\hbar\omega[0.693\coth(\hbar\omega/2kT)]^{1/2}, \qquad (1)$$

where $\hbar\omega$ is the energy of the lattice mode and α indicates the strength of the impurity-lattice coupling. Figure 3 shows the measured values of the half-width of the N_X luminescence band as a function of temperature for $x=0.75$ GaAs$_{1-x}$P$_x$:N. For convenience, we define an effective half-width as $W_{1/2}=E_p-E_{1/2}$ where E_p is the photon energy at the maximum of the band and $E_{1/2}$ is the photon energy at the half-power point on the low-energy side of the luminescence band. As shown in Fig. 3, the data are well described by Eq. (1) with $\hbar\omega=21$ meV.

The peak of the N_X luminescence band (Fig. 4) decreases rapidly with respect to the X band edge as the crystal composition x decreases[12,13,15] and clearly is of different origin than the higher-energy curve labeled N_Γ. The circular data points of Fig. 4 (see Table I) represent N-trap luminescence transitions that have been identified as the A line for[12,13] $0.40\lesssim x\lesssim 0.53$ and NN_3 pairs for $0.37\lesssim x\lesssim 0.38$.[15] In Fig. 5 we have reproduced data from Ref. 15 showing the emission spectra (77 °K) of N-doped and N-free GaAs$_{1-x}$P$_x$:N at composition $x=0.37$. While the N-free sample exhibits the expected band-to-band luminescence, *two* additional transitions, N_X and N_Γ, are observed for the N-doped sample. The transition labeled N_X corresponds to the N_X band of Figs. 1, 2, and 4. As mentioned above, previous work has attributed the N_Γ peak of Fig. 5 to NN_3 pairs.[15] Recent photoluminescence studies on N-implanted GaAs$_{1-x}$P$_x$ indicate, however, that the transition labeled N_Γ in Fig. 5 is due to an additional nitrogen bound state with a smaller binding energy than that of N_X.[16] We label this transition N_Γ in both the direct and indirect composition regions although its character changes beyond the direct-indirect transition where $E_X<E_\Gamma$.

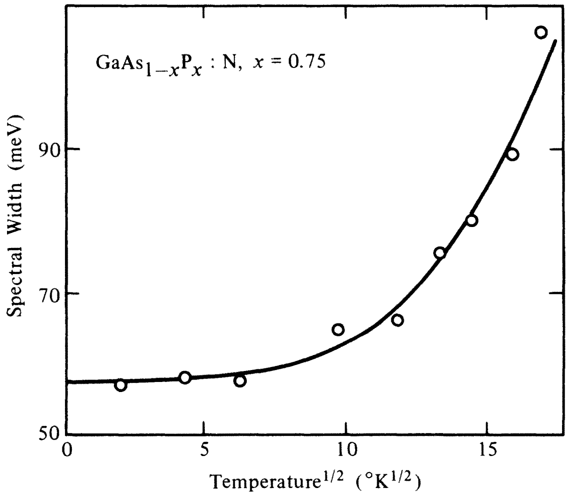

FIG. 3. Effective half-width $W_{1/2}$ (as defined in the text) for the N-trap luminescence ($x=0.75$ GaAs$_{1-x}$P$_x$:N) as a function of the square root of temperature. The solid curve is Eq. (1) adjusted to fit the experimental data ($\hbar\omega=21$ meV).

To investigate the behavior of this state, we have made pressure measurements on In$_{1-y}$Ga$_y$P$_{1-z}$As$_z$-GaAs$_{1-x}$P$_x$:N heterojunction diodes (the diode active region is p type and lies in the GaAs$_{1-x}$P$_x$ substrate) in the composition range $0.38\lesssim x\lesssim 0.48$. Figure 6 shows the pressure variation of the N_X, N_Γ, and the band edge (Γ) to acceptor (Zn) luminescence peaks of an $x=0.42$ diode.[18,25] The Γ-Zn transition shows the characteristic pressure behavior of the direct band gap.[21] Above ~4.5 kbar, the crystal is indirect as

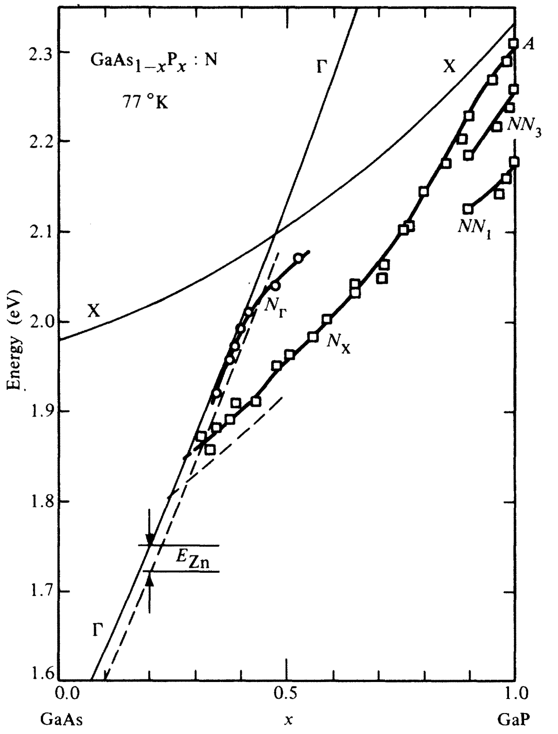

FIG. 4. Energies (77 °K) of the N_X band ("A line") and higher energy N_Γ state in GaAs$_{1-x}$P$_x$:N. Energies of the Γ and X conduction band minima are also shown. For $x\gtrsim 0.90$ NN_1 and NN_3 pairs are still distinguishable. Below $x\sim 0.90$, the NN pairs are not distinguishable and the A line of GaP:N becomes a broadened N_X band. The circular data points represent the photoluminescence peaks of a second N-trap level observed for $x\lesssim 0.55$.

TABLE I. Position of the N trap in $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ near the Γ and X band edges for crystal composition $0.35 \lesssim x \lesssim 0.53$. For reference the Γ and X band edges at 77 °K vary as (Ref. 12 and 26) $E_\Gamma(x) = 1.514 + 1.174x + 0.186x^2 E_x(x) = 1.977 + 0.144x + 0.211x^2$ and if the point at $x = 0.35$ is ignored or is actually of lower composition $E_N(x) \approx 0.993 + 3.934x - 3.613x^2$, $0.35 < x < 0.55$.

Crystal composition x	0.35	0.37	0.385	0.40	0.42	0.425	0.435	0.48	0.53
N-trap energy (eV)	1.918	1.951	1.968	1.990	2.010	2.015	2.019	2.040	2.065

indicated by the small negative pressure coefficient of the X transition; i.e., in the pressure range $\gtrsim 4.5$ kbar E_X is lower in energy than E_Γ. The N_Γ transition follows Γ for $p \lesssim 3$ kbar (corresponding to $x \lesssim 0.45 \approx x_c$) and bends and follows X for $p \gtrsim 5$ kbar. Also shown in Fig. 6 is the energy of the N_X band, which increases slightly for pressures to ~3 kbar, then decreases for $p \gtrsim 3$ kbar. We note that the pressure behavior of N_Γ and N_X of Fig. 6 (and also of diodes of composition $x = 0.38$ and 0.40) verifies, in a continuous manner, the position of the discrete data points of Fig. 4 ($x \lesssim 0.49$).

The behavior of the N trap (in $\mathrm{GaAs}_{1-x}\mathrm{P}_x$) at higher compositions ($x > x_c$) is revealed by the spectral behavior of the $x = 0.48$ diode of Fig. 7 as pressure is increased. Note that at composition $x \approx 0.48$ and 77 °K the diode active region is beyond the direct-indirect transition ($x_c \approx 0.45$, 77 °K),[26] and recombination from the Γ valley ($E_\Gamma - E_{\mathrm{Zn}}$ ~2.086 eV) is not observed. Rather, at lower pressures recombination involving N_X and N_Γ (cf. Fig. 4) is observed as shown in Fig. 7(a). We mention that this emission agrees with Zn-diffused junctions prepared on the same substrate.[12] The ~+16 meV shift to higher energy observed between curves (a) and (b) for both the N_X and N_Γ transitions (Fig. 7) may be attributed to the decreasing effect of the Zn acceptor in the recombination process as the crystal becomes more indirect with increasing pressure.[4,5] This is an interesting impurity effect in its own right that is sensitive to the crossover from direct to indirect gap.[4,5]

Of primary interest here, however, is the variation in intensity of the state labeled N_Γ as pressure

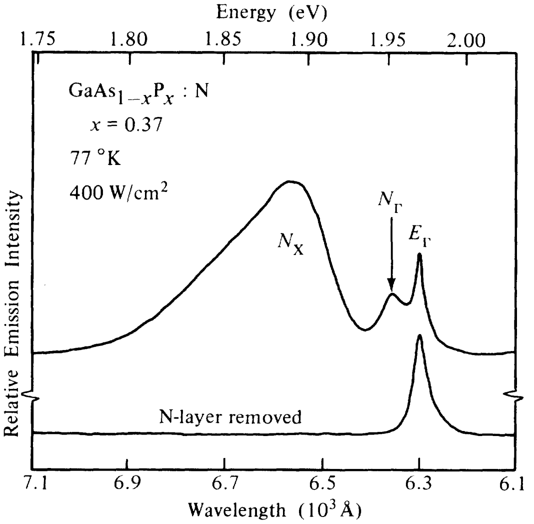

FIG. 5. Photoluminescence spectra (77 °K) of N-free and N-doped $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ ($x = 0.37$). Band-to-band recombination is observed in the N-free sample. The N-doped sample shows the N_X and N_Γ states expected for $x < 0.55$.

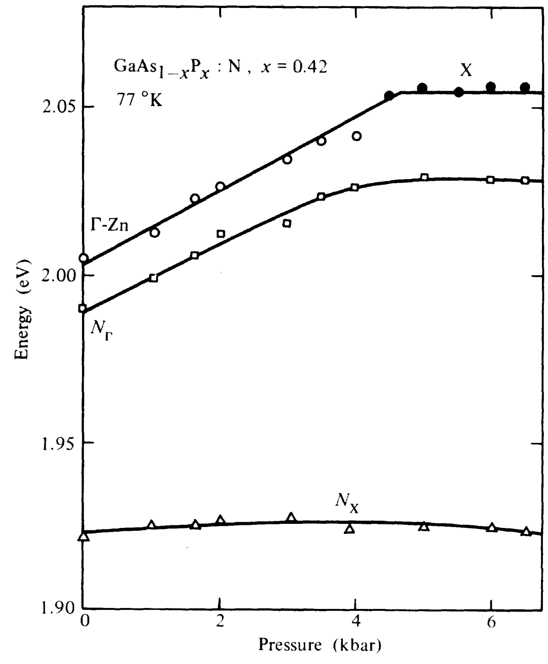

FIG. 6. Pressure variation of N_X, N_Γ, and Γ-Zn transitions of an $\mathrm{In}_{1-y}\mathrm{Ga}_y\mathrm{P}_{1-z}\mathrm{As}_z$-$\mathrm{GaAs}_{1-x}\mathrm{P}_x$:N ($x = 0.42$) single heterojunction. For $p \gtrsim 4.5$ kbar the crystal becomes indirect and the Γ-Zn transition is replaced by that involving the X band edge (and its associated pressure behavior). The N_Γ state follows the Γ band edge to p ~3 kbar (x ~0.45) and the X-band edge for $p \gtrsim 5$ kbar ($x \gtrsim 0.47$). The energy of the N_X band is observed to increase slightly as the direct-indirect crossover is approached.

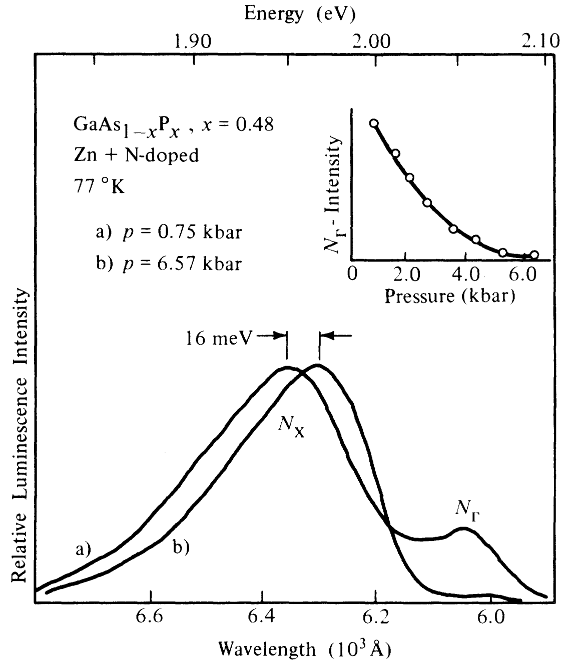

FIG. 7. Spectral behavior (77 °K) and N_Γ-level intensity variation with pressure (inset) of an $In_{1-y}Ga_yP_{1-z}As_z$-$GaAs_{1-x}P_x$:N ($x=0.48$) single heterojunction (constant current density, $J=6\times10^3$ A/cm^2). The N_Γ-level luminescence intensity (inset) decreases with pressure owing to the rapid increase in energy of the Γ band edge, and increase in $E_\Gamma-E_N$, and thus decrease of the $k=0$ wavefunction component of the N_Γ-level bound electron. Curve b is shifted (~16 meV) to higher energy because of the decreased effect of the Zn acceptor as the crystal becomes more indirect with increased pressure.

is increased (see inset of Fig. 7, diode current constant at $J=6\times10^3$ A/cm^2). At low pressures, $E_\Gamma - E_N \approx 70$ meV, and the N_Γ transition is clearly observed. As the pressure is increased and $E_\Gamma - E_N$ increases (to ~140 meV at 6.57 kb), the intensity of the N_Γ transition is observed to decrease relative to that of N_X. This is in agreement with the circular data points of Fig. 4 which indicate that the transition labeled N_Γ has not been observed at crystal compositions $x>0.53$.[27]

DISCUSSION

The pressure data described here for $GaAs_{1-x}P_x$:N ($0.38 \lesssim x \lesssim 0.48$) have demonstrated in a continuous manner the behavior of the two nitrogen states N_Γ and N_X in the vicinity of the direct-indirect crossover ($x=x_c\approx 0.45$, 77 °K).[26] This work agrees with recent photoluminescence studies on N-implanted $GaAs_{1-x}P_x$ (Refs. 10 and 16) which indicate that the states previously identified as NN_3 at composition $0.37\lesssim x\lesssim 0.38$[15] and the A line in the range $0.40\lesssim x \lesssim 0.53$[12,13] are actually the same N-impurity level, a mixture of Γ and X components near crossover. The decreased oscillator strength observed here for this state (N_Γ) as $E_\Gamma - E_N$ increases with pressure may be attributed to a lessening of the $k=0$ component of the bound-electron wave function as the crystal becomes more indirect. Although not shown in the data of Fig. 7, we note that the original experimental data indicate that as the N_Γ-state transition decreases in intensity, it becomes indistinguishable from the Te_0 - LA (or E_{gx} - LA)[12] transition for $p \gtrsim 5$ kbar ($x\gtrsim 0.54$).

It is interesting to note that the N_Γ state and transition described here has sufficient oscillator strength in the range $x=0.37$–0.38 so that it can be photopumped on the N_Γ state itself (i.e., $E_N < E_p < E_\Gamma$) and be operated in stimulated emission.[28] This is now understandable. In the range $0.38 \lesssim x \lesssim 0.43$ this state has been operated in stimulated emission in heterojunctions but at *higher* current thresholds than in the *same* diode structures free of N doping.[8,25] That is, the N_Γ state to some extent hinders band-to-band recombination. Nevertheless, in the even more difficult range $x_c<x<0.47$ (in accord with the N_Γ emission of Fig. 7), this state has operated in stimulated emission by either photopumping[29] or electron beam pumping[27] (but not in diodes). In view of the present work (Fig. 7), probably the upper composition limit for *laser* emission in $GaAs_{1-x}P_x$:N has not yet been attained (but most likely does not exceed $x \sim 0.55$).

Of further interest, and now understandable in view of Refs. 10, 16, and the present work, the N_X state (transition) has recently been extended in laser operation from[11] $x=0.35$ to $x=0.37$,[30] i.e., to a region of weaker band-structure enhancement[3] (*but still direct-gap crystal*). Just as for the case of the N_Γ state, in heterojunctions the N_X state has been observed to hinder or weaken radiative recombination in comparison with band-to-band recombination in otherwise similar N-free diodes.[31] Note that the results we have described here do not speak well for the case for stimulated emission in GaP:N. Finally, we remark that for the N_X state the extension of stimulated emission to higher crystal composition is not nearly as important as the use of this state in light-emitting diodes, which are optimum at $x\sim 0.65 > x_c$.[1-3]

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