Luminescence decays of N-bound excitons in $GaAs_{1-x}P_x$

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(Received 20 July 1982)

We report photoluminescence-lifetime studies of nitrogen-bound excitons in $GaAs_{1-x}P_x$:N at low excitation power for $0.22 < x \leq 1.0$. Temperature was varied from $T=1.8$ to 200 K. A time resolution of 400 psec was obtained with the use of time-correlated single-photon counting. At low temperatures, a short (<10 nsec) nonexponential decay and a longer matrix-element-determined exponential decay are observed. Results prove that earlier 77-K phase-shift measurements gave information regarding only the nonexponential component. We report here on the true exponential luminescence lifetime. Luminescence-lifetime quenching as the temperature is raised from 1.8 to 40 K demonstrates that both $A(J=1)$ and $B(J=2)$ bound-exciton states exist in alloys with minimal strain mixing. Radiative lifetimes of both exciton spin states are obtained across the alloy for the first time and are compared with earlier theories. Both lifetimes show a band-structure enhancement of 2 orders of magnitude, with A-state radiative lifetimes varying from about 100 nsec in GaP to 1 nsec for $x=0.23$. When the bound exciton is near resonance with the direct edge (for $x \cong 0.22$), the radiative lifetime approaches but does not fall below the direct-edge luminescence lifetime, thus showing that the recombination rate does not diverge with resonance. For $T > 40$ K, two distinct thermal processes occur, one of which (hole release) increases the luminescence lifetime, while the other (electron detrapping) decreases the lifetime and quenches the luminescence intensity. Observation of hole release confirms the Hopfield-Thomas-Lynch model of N isoelectronic traps, and proves for the first time that isolated N binds a bare electron.

I. INTRODUCTION

Lifetimes of excitons bound to N traps in $GaAs_{1-x}P_x$:N alloys (N_X excitons) have been investigated both experimentally and theoretically. In particular, a decrease (of orders of magnitude) in the N_X radiative lifetime has been predicted[1,2] under conditions where the bound-exciton level becomes resonant with the direct conduction-band edge. Although Chevallier *et al.*[3] observed a decrease of a factor of 4 in the N_X luminescence lifetime at 4.2 K as X varied from 1.0 (GaP) to 0.59, the complete band-structure enhancement as the N_X level approaches degeneracy with the direct edge has never been fully confirmed experimentally. This band-structure enhancement[1] (BSE) occurs because the modulus of the wave function of the trapped electron near $\vec{k}=0$ in the Brillouin zone increases in the presence of the direct conduction-band edge. The hole wave function is always concentrated near $\vec{k}=0$; thus the exciton recombination rate increases due to the increased electron-hole overlap in $\vec{k}$ space. The reason for an order-of-magnitude enhancement across the $GaAs_{1-x}P_x$ alloy is that the energy separation between the direct conduction-band edge and the N_X level varies[5] from 0.5 eV in GaP to zero for $x \cong 0.22$, as may be inferred from the alloy diagram in Fig. 1. (A calculation[2] of the enhancement is shown in Fig. 9.) Previous lifetime measurements[3,4] have covered only a limited range of the alloy, and each report was for a single temperature. In addition, Chevallier *et al.*[3] ($x \geq 0.59$, $T=4.2$ K) used a high-power nitrogen laser source (although no heating effects were observed). The work of Lee *et al.* and Nelson *et al.*[4] ($x \geq 0.35$, $T=77$ K) used the phase-shift method with a 140-MHz mode-locked laser pulse train, and therefore assumed simple exponential decays with decay times limited to $\lesssim 10$ nsec.

This work presents a study of the actual bound-exciton lifetime throughout the entire range of the alloy where the N_X state of isolated N lies within

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the energy gap ($x>0.22$).[5] In addition, temperature dependence of the N_X lifetime was measured from 1.8 to 200 K. Such a temperature study was necessary to understand the lifetime results because of various thermal processes which occur. The experimental technique used was time-correlated single-photon counting,[6] which allowed direct measurement of luminescence lifetimes greater than 400 psec. Our direct observation of luminescence decays shows significant deviations from single-exponential behavior at short times ($\lesssim 20$ nsec). Previous phase-shift measurements resolved only this nonexponential part and were unable to measure the longer radiative (i.e., matrix-element-determined) lifetime. The nonexponential decay, which we believe results from exciton diffusion within the alloy-broadened N_X band, will be the subject of a later paper.[7]

Our results for low temperatures (<40 K) verify the band-structure enhancement in the transition matrix element: The matrix element increases by about 2 orders of magnitude as x is varied from $x=1.0$ (GaP) to $x=0.23$ (where the exciton level becomes degenerate with the direct-band edge). This

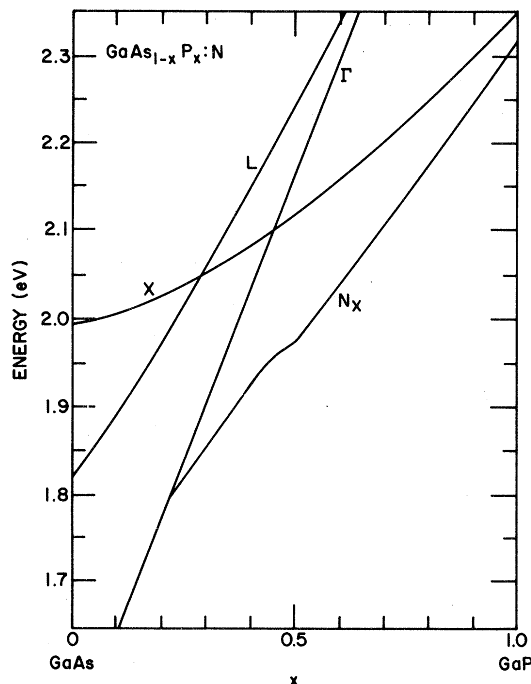

FIG. 1. Energy diagram for the nitrogen exciton N_X level in $GaAs_{1-x}P_x$:N alloys at 5 K. Γ is the direct ($\vec{k}=0$) conduction-edge minimum, X is the [100] minimum, and L is the [111] minimum. The energy reference is the valence-band maximum. The binding energy of the exciton is the smaller of $E_\Gamma - E_{N_X}$ and $E_X - E_{N_X}$, and is largest around $x=0.45$. (After Ref. 5.)

enhancement is in qualitative agreement with the calculation of Kleiman.[2] The low-temperature lifetime dependence also confirms that distinct A ($J=1$) and B ($J=2$) states of the bound exciton exist throughout the alloy. These levels, obscured by inhomogeneous alloy broadening in ordinary photoluminescence spectra, have been reported in resonant excitation experiments.[8] Despite alloy-induced strain, we find the A-state transition matrix element remains much larger than that of the B state throughout the alloy.

At temperatures greater than 40 K, two distinct thermal detrapping processes become important. For deeply bound N_X excitons ($0.4 \lesssim x \lesssim 0.6$) and temperatures up to about 100 K, the excitonic hole is thermally released, leaving behind a trapped electron. This hole release verifies the Hopfield-Thomas-Lynch (HTL) model[9] of N traps and recent theoretical models[10] in which the central-cell potential is believed to be electron-attractive and responsible for binding. The binding of a bare electron by isolated N has been questioned in several papers,[11,12] and, while near-neighbor nitrogen pairs in GaP have been shown to bind electrons,[13,14] this is the first report for isolated N. For other alloy compositions, and for temperatures greater than 100 K for deeply bound N_X excitons, hole and electron are both detrapped, with resulting recombination of the pair at the band gap.

II. EXPERIMENTAL

The principal method used in this study was time-correlated single-photon counting.[6] The setup is shown in Fig. 2. The laser source was a cw mode-locked argon laser operating at 5145 Å (above the band gap throughout the alloy). Pulses from the 70-MHz train were acousto-optically selected at a convenient repetition rate (typically 350 kHz). The switched 150-psec pulses contained about 1 nJ of energy. Half of each switched pulse was focused onto 150-psec risetime photodiode; the remainder was

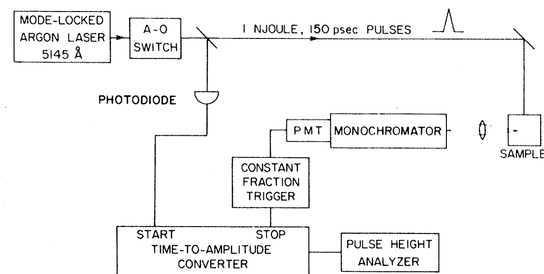

FIG. 2. Block diagram of time-correlated single-photon counting apparatus.

focused onto the sample with a spot size of about 100 μm. Samples, mounted to copper heatsinks, were placed in a Janis immersion Dewar for the 1.8-K measurements, and a Janis Super Varitemp Dewar equipped with a silicon diode temperature sensor and controller for the other measurements. Sample luminescence was collected through a Spex 1401 double monochromator and focused onto an RCA C31034 photomultiplier. The single-photon pulses from the phototube were amplified and sent to a constant fraction trigger. The trigger produced the stop pulse for a time-to-amplitude converter (TAC). The start pulse for the TAC was the photodiode output. The varying-amplitude pulses from the TAC were collected as a histogram on a pulse-height analyzer. Provided the photomultiplier count rate was much less than the pulse repetition rate, the histogram built up over several minutes gave the luminescence decay directly. As shown in Fig. 3, the response of the system at the laser wavelength was a 400-psec-wide [full width at half maximum (FWHM)] histogram, which sets the system resolution. This resolution is limited by the transit-time dispersion in the photomultiplier.

With the laser pulse energy of 1 nJ, no heating effects were observed when varying either pulse repetition rate or pulse energy. To link our results with the previous work of Chevallier *et al.*,[3] luminescence decays were measured at 4.2 K using 10-kW 10-nsec pulses from a nitrogen laser ($\lambda = 3371$ Å). This emission was collected through the monochromator and phototube. The phototube signal was averaged with a boxcar integrator. The resolution limit of this system was the 10-nsec laser pulse width.

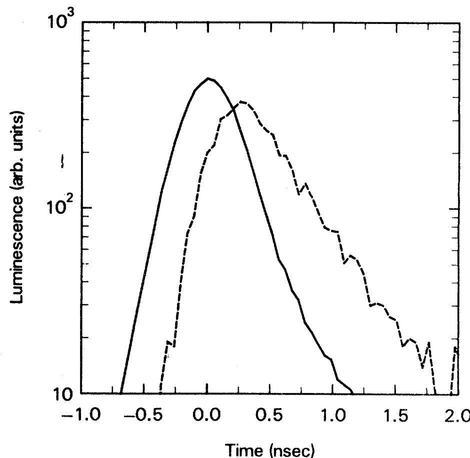

FIG. 3. Response (solid line) of the time-correlated photon-counting system to 150-psec 5145-Å pulses. For reference, a 430-psec decay (dashed line) measured with the system is also shown.

The $GaAs_{1-x}P_x$ crystals used in this work were prepared by conventional halide-transport vapor-phase epitaxial (VPE) techniques.[15] Samples were prepared on bulk (100) GaP substrates for $x \gtrsim 0.5$, and on (100) GaAs for $x \lesssim 0.5$. Compositional grading between the substrate and the final layer was used to reduce misfit dislocations. The epitaxial layers were ~ 50 μm thick and either nominally undoped ($\sim 10^{15}$ cm^{-3} *n*-type) or intentionally Te doped ($\sim 10^{17}$ cm^{-3}). The final 10–20 μm of growth were N-doped by the addition of NH_3 to the H_2 carrier gas. Incorporated nitrogen concentrations are estimated to be in the range 10^{17}–10^{18} cm^{-3}, as deduced from comparisons of the photoluminescence spectra of these VPE samples to the spectra of nitrogen-implanted samples.[16]

III. TEMPERATURE STUDY ON ONE SAMPLE

In this section, detailed lifetime measurements on a particular sample of composition $x = 0.39$ will be discussed. Systematics of the results across the alloy are considered in the next section. Figure 4 shows the luminescence decay of the sample at the peak (1.91 eV) of the N_X luminescence at various temperatures. We note two distinct regimes, which become especially clear at low temperatures. At the shortest times there is a rapid ($\lesssim 10$ nsec) nonexponential decay of the luminescence intensity. This decay, which will be the subject of a later paper,[7] results from exciton diffusion within the alloy-broadened impurity band. The exponential decay

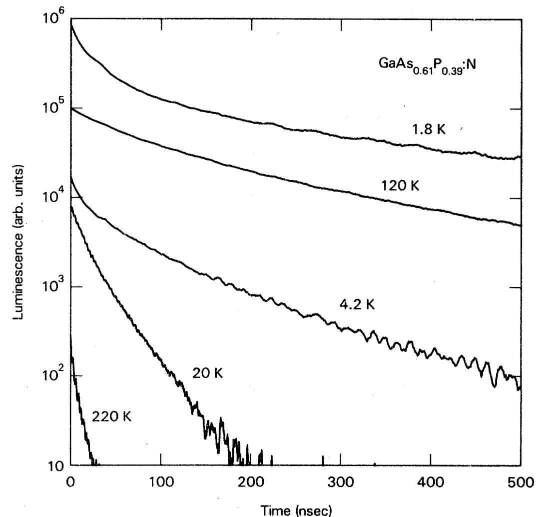

FIG. 4. Bound-exciton luminescence decays at various temperatures at the luminescence peak of a sample with alloy composition $x = 0.39$. The amplitudes have been scaled simply to separate the curves. The nonexponential short-time decay is especially evident in the 1.8-K curve.

which appears at longer times represents the bound-exciton decay lifetime τ_d. The exciton luminescence band has a FWHM $\cong 80$ meV at this alloy composition, and we find that τ_d increases with exciton binding energy typically $\sim 20\%$ across the exciton band, as shown in Fig. 5. This increase may be due to both a decreased BSE and to residual transfer within the band. Our results disagree substantially with the phase-shift measurements of Lee *et al.* and Nelson *et al.*,[4] which suggested a lifetime varying strongly across the band and always less than 10 nsec at 77 K. We believe that these earlier measurements resulted from the fast nonexponential transfer and do not represent the N_X radiative lifetime.

The lifetime at the N_X luminescence peak as a function of temperature for this sample is shown in Fig. 6(a). The lifetime curve can be separated into three regions:

Region 1. For temperatures less than 40 K we find a strong monotonic quenching of τ_d with temperature which begins at the lowest T. This result agrees with the well-known temperature variation for isoelectronic traps,[17] and specifically N in GaP. The quenching results from thermalization between the A ($J=1$) and the lower-energy B ($J=2$) state with the N_X band. From these two distinct exciton spin states at each N site with respective radiative lifetimes τ_A and τ_B, and separated by an energy $E_A - E_B = \Delta_{AB}$, the observed decay time should vary according to the expression[17]

$$\tau_d = \frac{\tau_B[1 - g\exp(-\Delta_{AB}/kT)]}{[1 - \gamma g \exp(-\Delta_{AB}/kT)]}, \qquad (1)$$

where g is the ratio of the degeneracies of states A and B, and $\gamma = \tau_B/\tau_A$. This expression is valid provided the thermalization time between the levels is much less than both τ_A and τ_B, and no other relaxation processes are important. We could not observe any difference in the decay kinetics of the A and B lines in GaP, so the thermalization time must be short in GaP. We will assume it is also short in $GaAs_{1-x}P_x$. In alloys, because the N_X luminescence is an inhomogeneously broadened band, the A and B levels cannot be resolved in conventional photoluminescence spectra. Nonetheless, they have been detected as a discrete substructure with the alloy-broadened N_X band in resonant excitation spectra.[8] Supporting the observation, we can apply Eq. (1) in the alloy, giving the solid theoretical curve of Fig. 6(a), obtained for $\Delta_{AB} = 0.8$ meV, $g = 0.6$, $\tau_B = 550$ nsec, and $\tau_A = 9.6$ nsec. In this alloy, however, Δ_{AB} may not represent the splitting $E_A - E_B = 5$ meV previously deduced from resonant excitation.[8] If an exciton can transfer from the B level on one nitrogen site to the A level on another site, then Δ_{AB} may represent the activation energy for this process. As seen in Fig. 6(a), the data clearly do not fit a curve for $\Delta_{AB} = E_A - E_B$. In any case, the fitted values τ_A and τ_B should correspond to actual A- and B-state radiative lifetimes. In the limit $kT \ll \Delta_{AB}$, all the excitons will be in the lower-energy B state, so that $\tau_d = \tau_B$ as in Eq. (1). For $kT \gg \Delta_{AB}$, all states are equally populated, so that, again as in Eq. (1) $\tau_d = \tau_B(1-g)/(1-\gamma g)$. The assumption that $g = 0.6$ throughout the alloy is reasonable provided

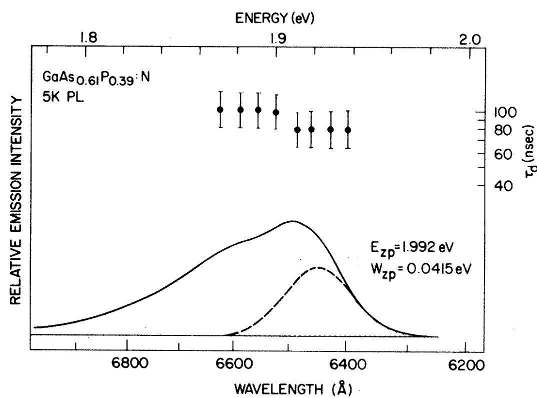

FIG. 5. Time-integrated luminescence-intensity spectrum (solid line, below) for the sample of Fig. 4. The dashed line is an approximate fit to the actual N_X luminescence line obtained assuming linear phonon coupling as in Ref. 8. E_{zp} and W_{zp} are the peak energy and width (FWHM) of the fitted zero-phonon line. The data above give the luminescence lifetime measured at 5 K vs photon energy.

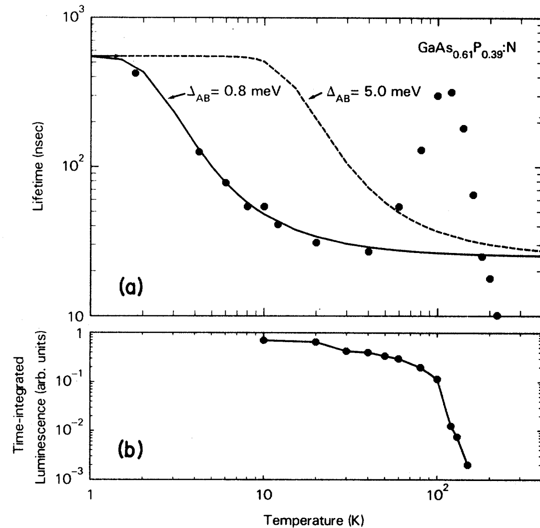

FIG. 6. (a) N_X luminescence lifetime vs temperature for the same sample as in Fig. 4. The curves are plots of Eq. (1) with $\tau_B = 550$ nsec, $\tau_A = 9.6$ nsec, $g = 0.6$, and Δ_{AB} as indicated. (b) Time-integrated luminescence at the bound-exciton peak vs temperature for the same sample.

the $J=1$ and $J=2$ states are still largely unmixed. Our deduced τ_A and τ_B are physically reasonable for allowed A- and forbidden B-state transitions associated with each N site, confirming that the analog of the A and B states of isolate N in GaP are still largely unmixed in $GaAs_{1-x}P_x$. The existence of separate A and B states on a single site in the alloy requires that the N_X exciton energy on a single site be well defined. However, the exciton energy varies from site to site, so that the exciton band is inhomogeneously broadened by alloying.[8,18]

Region 2. As the temperature is raised above 40 K, other thermal processes begin to dominate the measured lifetimes. For the region from 40 to 100 K, the luminescence lifetime increases by a factor of 50. Cuthbert and Thomas have previously observed this behavior for excitons bound to O isoelectronic traps in ZnTe.[17]

In the HTL model[9] of the isoelectronic N bound exciton, the electron is bound to the short-range central-cell potential of the nitrogen impurity, and the hole is then bound in the Coulomb field of the electron. If the electron is bound tightly to the nitrogen site, then it will be possible to thermally release the hole, leaving the electron still bound to the impurity. The hole remains in the valence band until it is retrapped on the same (or another) bound electron. As a result, the hole is separated from the trapped electron for a large fraction of its lifetime, so the measured luminescence decay time will increase proportionately. Provided there are no other recombination processes, the time-integrated luminescence intensity will not change significantly in this region, as seen in Fig. 6(b).

Region 3. Above 100 K, the luminescence lifetime falls rapidly. Thermal energy is now sufficient to detrap both hole and electron, which now recombine radiatively or nonradiatively at the band gap. The time-integrated exciton luminescence intensity thus falls rapidly with temperature [Fig. 6(b)], while the N_X luminescence decay time approaches that of the band-edge processes. (We find the band-gap luminescence lifetime remains constant at about 0.5 nsec as the temperature is varied.) Thus, at high temperatures recombination kinetics are controlled by band-gap recombination.

IV. SYSTEMATICS ACROSS THE ALLOY

Measured luminescence decays for various alloy compositions at 4.2 K are presented in Fig. 7, which illustrates the large BSE across the alloy. Luminescence lifetimes of bound excitons versus temperature for a series of alloy compositions are shown in Fig. 8. We note that the region-1 behavior is similar throughout the alloy, and we may extrapolate A- and B-state lifetimes as explained in the previous section. These extrapolated lifetimes, along with the measured luminescence lifetimes at 4.2 K, are shown in Fig. 9. The theoretical calculation is due to Kleiman.[2] We have corrected the exciton binding energy from that previously assumed to the recent data for the N_X exciton,[5] (shown in Fig. 1) and also corrected the $x=1.0$ (GaP) radiative lifetime. We assume

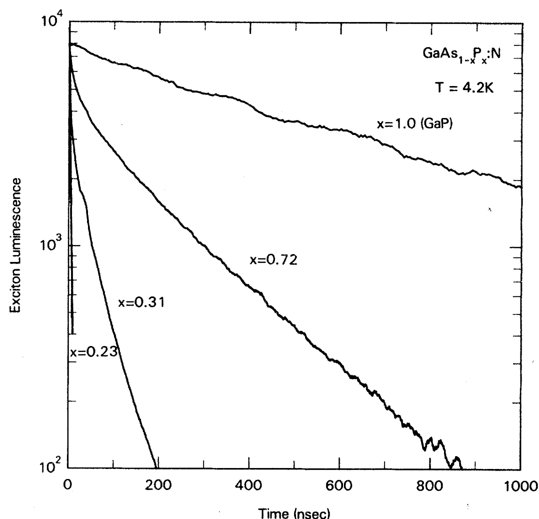

FIG. 7. N_X luminescence intensity decays for a series of alloy compositions at 4.2 K. Amplitudes have been scaled to separate the curves.

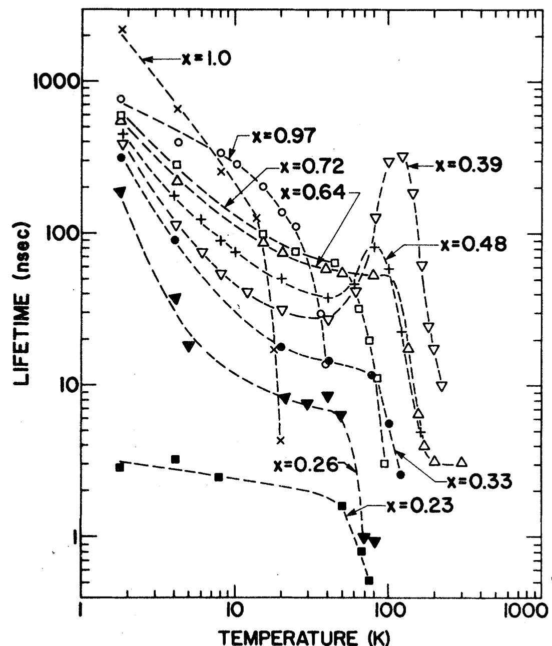

FIG. 8. N_X luminescence lifetime vs temperature for a series of alloy compositions. Dashed lines are drawn through the data for clarity.

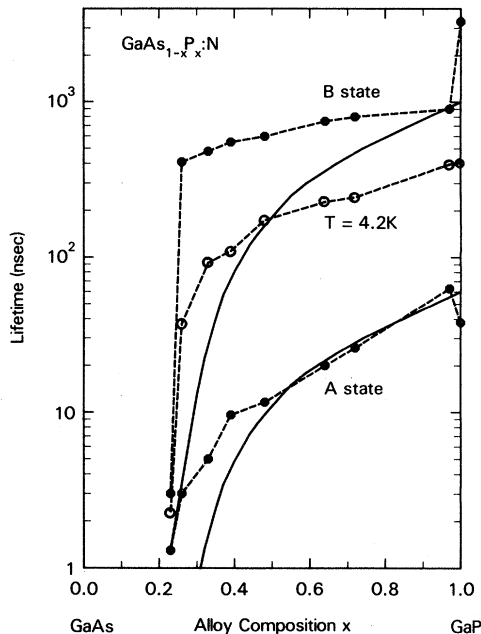

FIG. 9. Extrapolated A- and B-state lifetimes (filled circles) and measured 4.2-K N_X lifetime (open circles) vs alloy composition. Data are connected by dashed lines for clarity. Solid curves are from the corrected theory of Ref. 2, as explained in the text.

that the calculation is valid for both A and B states. This assumption implies that A and B states on a given nitrogen site are not mixed strongly by microscopic alloy strain or misfit-induced strain; the deduced relation $\tau_B/\tau_A \gg 1$ confirms this assumption except possibly for $x < 0.25$. The lifetimes of both A and B states decrease by about 2 orders of magnitude as the alloy composition is varied from $x = 1.0$ to $x = 0.22$. This band-structure enhancement is in general agreement with the theory. In no case is the N_X radiative lifetime shorter than the band-gap luminescence lifetime. Thus the band-gap recombination seems to pose a limit on the N_X radiative rate, in contrast to some earlier suggestions.[1,19]

As seen in Fig. 8, the measured effective activation energy [Δ_{AB} in Eq. (1)] remains roughly constant throughout the alloy with value $\Delta_{AB} \cong 0.8$ meV in our samples. The dependence of Δ_{AB} on nitrogen concentration has been investigated in a series of nitrogen-implanted samples[16] (implanation depth $\cong 3500$ Å) with alloy composition $x = 0.46$. No variation in Δ_{AB} was seen as the concentration was varied from 10^{16} to 10^{18} atom/cm^{-3}. This result is perhaps not surprising for the thermally activated process discussed in the previous section, in contrast to a tunneling process which should be concentration dependent. However, the fact that the measured value of Δ_{AB} throughout the alloy corresponds to $E_A - E_B$ in GaP may not be simply fortuitous, and further investigations continue.

The results of our measurements at $T = 4.2$ K are in reasonable agreement with the previous results of Chevallier *et al.*[3] The lifetimes we report are 10–20 % longer than the previous results. Measuring luminescence lifetimes with a nitrogen laser as described in Sec. II gave results nearly identical with those of Chevallier *et al.* when the nitrogen laser pulse was attenuated to about 1-W peak power. For laser powers of 10 kW, lifetimes dropped by about a factor of 2, corresponding to a temperature increase of about 5 K.

Region 2, where the lifetimes increase with temperature, is found only over a limited range of the alloy, $0.4 \lesssim x \lesssim 0.6$. Assuming the hole is trapped in the Coulomb field of the electron, the hole binding energy remains roughly constant at about 40 meV (Ref. 3) throughout the alloy. The electron binding energy to the nitrogen site, on the other hand, must then vary strongly across the alloy. Only where the electron binding energy is large can the hole be released separately, leaving the electron still trapped. Large binding energies occur just where region 2 is observed (compare with Fig. 1). This observation of hole release demonstrates that the electron is the principal particle bound to N (as in the HTL model) and that its separate thermalization to the band edge controls ultimate quenching of the N_X exciton luminescence.

From an Arrhenius plot of $\ln\tau_d$ against T^{-1} in region 2, it is possible to deduce an approximate value for the hole binding energy E_H. For the data in Fig. 6, we estimate $E_H \sim 30$ meV. This value further confirms the THL model, as it is characteristic of the 40-meV hole binding energy[13] in excitons bound to near-neighbor nitrogen pairs in GaP. Our measured binding energy is much larger than the ~ 5-meV binding energy found for free direct-gap excitons at this alloy composition,[20] thus showing that the hole is bound to a well-localized electron.

The observation of hole release only shows that isolated N can bind a bare electron for alloy compositions $0.4 \lesssim x \lesssim 0.6$. However, near-neighbor nitrogen pairs in GaP have been shown to bind a bare electron[13] and, under pressure, the N_X exciton level has been observed to emerge from the direct-band edge in GaAs.[21] Taking all these observations together, it seems very likely that the HTL model[9] is valid throughout the alloy in that the interaction responsible for binding the exciton to isolated N is between the electron and N alone via the central-cell potential.[10] The previous suggestion that the exciton binds because of an interaction between the entire exciton and N through the impurity-induced strain field[11] is not borne out by these observations.

At higher temperatures, both time-integrated luminescence intensity and luminescence lifetime decrease sharply in all alloys as both holes and electrons are thermally detrapped. Activation energies deduced from Arrhenius plots of $\ln\tau_d$ vs T^{-1} in this temperature region correlate well with exciton binding energies shown in Fig. 1. Arrhenius plots[14,22] of $\ln I$ against T^{-1} where I is the time-integrated luminescence also correlate well with binding energy. Furthermore, in direct-gap materials, we can measure the free carrier lifetime directly by measuring the band-edge luminescence lifetime. At high temperatures, we find that the exciton lifetime approaches the free-carrier lifetime. (Experimentally, the free-carrier lifetime is nearly independent of temperature.) Thus, the exciton lifetimes in all these alloys are limited by free-carrier recombination at high temperatures.

V. CONCLUSIONS

We have measured A- ($J=1$) and B- ($J=2$) state radiative lifetimes of excitons bound to N traps in $GaAs_{1-x}P_x$ alloys for $0.22<x\leq 1.0$, thus covering for the first time the entire range of the stable exciton bound to nitrogen. Both lifetimes decrease by 2 orders of magnitude as x decreases, which agrees with earlier calculations of the band-structure enhancement. A- and B-state lifetimes remain significantly different throughout the alloy, pointing out that despite alloy-induced strain, allowed A and forbidden B-state transition are associated with each N site. The thermal activation to A states from B states as the temperature is increased to 40 K remains to be fully characterized. As the temperature is raised from 40 to 100 K, luminescence lifetime increases for those alloy compositions in which the electron binding energy is large. The lifetime increase provides further support for the Hopfield-Thomas-Lynch model[9] of isoelectronic traps, as it is due to thermal release of the hole, leaving the electron still trapped. At sufficiently high temperatures, both hole and electron are detrapped, so that the luminescence intensity is quenched and the exciton lifetime decreases to that of the free carriers.

ACKNOWLEDGMENTS

This work was supported in part by the Office of Naval Research under Contract No. N00014-80-C-0376.

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