Unusual carrier thermalization in a dilute Ga As $1 - x$ N $x$ alloy

P. H. Tan, Z. Y. Xu, X. D. Luo, W. K. Ge, Y. Zhang, A. Mascarenhas, H. P. Xin, and C. W. Tu

Citation: Applied Physics Letters 90, 061905 (2007); doi: 10.1063/1.2454552
View online: http://dx.doi.org/10.1063/1.2454552
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/90/6?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in

Group-III intermixing in In As/In Ga Al As quantum dots-in-well

Quantum well intermixing for the fabrication of InGaAsN/GaAs lasers with pulsed anodic oxidation
J. Appl. Phys. 95, 3422 (2004); 10.1063/1.1651322

Carrier–carrier interaction in single In 0.5 Ga 0.5 As quantum dots at room temperature investigated by near-field scanning optical microscope

Gas-source molecular beam epitaxy of GaInNP/GaAs and a study of its band lineup
J. Vac. Sci. Technol. B 19, 1413 (2001); 10.1116/1.1381069

Mechanism for low-temperature photoluminescence in GaNAs/GaAs structures grown by molecular-beam epitaxy
Unusual carrier thermalization in a dilute GaAs$_{1-x}$N$_x$ alloy

P. H. Tan$^a$ and Z. Y. Xu
State Key Laboratory for Superlattices and Microstructures, Institute of Semiconductors, Beijing 100083, China

X. D. Luo and W. K. Ge
Department of Physics, HongKong University of Science and Technology, HongKong, China

Y. Zhang$^b$ and A. Mascarenhas
National Renewable Energy Laboratory, 1617 Cole Boulevard, Golden, Colorado 80401

H. P. Xin and C. W. Tu
Department of Electrical and Computer Engineering, University of California at San Diego, La Jolla, California 92039

(Received 11 October 2006; accepted 9 January 2007; published online 6 February 2007)

Photoluminescence (PL) properties of the $E_0$, $E_0 + \Delta_0$, and $E_s$ bands in an $x=0.62\%$ GaAs$_{1-x}$N$_x$ alloy were investigated in detail, including their peak position, linewidth, and line shape dependences on the excitation energy, excitation power, and temperature, using micro-PL. The hot electrons within the $E_s$ band are found to exhibit highly unusual thermalization, which results in a large blueshift in its PL peak energy by $>2k_BT$, suggesting peculiar density of states and carrier dynamics of the $E_s$ band. © 2007 American Institute of Physics. [DOI: 10.1063/1.2454552]

The giant band gap bowing in dilute GaAs$_{1-x}$N$_x$ alloy is a subject of intensive experimental and theoretical research in recent years because of its promising application in the optoelectronic fields and its peculiar fundamental physical properties. In addition to the unexpected large reduction of the fundamental band gap even with small amount of N incorporation, an electronic transition far above the band gap $E_0$, known as the $E_s$ transition, has been detected in several measurements, such as modulation reflectance (MR), resonant Raman scattering, and microphotoluminescence ($\mu$-PL). With increase of the nitrogen concentration, contrasting to the redshift of $E_0$, $E_s$ is found to blueshift with a magnitude of around 2/3 of that for $E_0$. It is well known that the PL spectrum depends strongly on the thermal distribution of carriers, and has been widely used to characterize the optical properties of the band gap emissions in Ga(In)NAs alloys. Recently, the deeply resonant $E_s$ band has been detected by $\mu$-PL at low temperature (80 K) in dilute GaAs$_{1-x}$N$_x$ alloys with $x$ as low as 0.1%, and the behavior of $E_s$ is found to be very different from another resonant transition $E_0 + \Delta_0$. The intrinsic optical transitions can be more clearly investigated with the $\mu$-PL technique because of the saturation of localized states by high excitation density. Although the broadening parameter of $E_s$ in MR is only 6% on average larger than those of $E_0$ and $E_0 + \Delta_0$ over a wide concentration range from MR measurements, the PL bandwidth of $E_s$ remains about 70 meV for $x$ being as low as 0.1%, which is much greater than that (25 meV) of $E_0$. It has been concluded that $E_s$ is originated from a rather large set of perturbed host states of GaAs near the L point. In this letter, using $\mu$-PL and a sample with $x=0.62\%$, we provide a comprehensive study on the temperature dependence of the $E_s$ transition between 80 and 300 K. The result shows that the $E_s$ band exhibits very unusual density of states and carrier thermalization compared to $E_0$ and $E_0 + \Delta_0$, which results in an unusually large thermalization-induced blueshift of its PL peak energy.

The GaAs$_{1-x}$N$_x$ sample ($x=0.62\%$) investigated here was grown by a gas-source molecular beam epitaxy on semi-insulating (001) GaAs substrates with an epilayer thickness of 400 nm. The detailed growth process has been described elsewhere. The $\mu$-PL is measured by a $\mu$-Raman system of Dilor Super Labram, which consists of a LN$_2$ cooled Si charge-coupled detector. The laser excitation energies are 1.959 eV of a He–Ne laser, 2.410 and 2.541 eV of an Ar$^+$ laser, and 2.089 and 1.848 eV of two diode pumped solid-state lasers. In order to avoid the strong interference of the Raman lines to the PL peak of $E_s$, a forbidden scattering configuration for longitudinal optical phonons at $\Gamma$ point was used for the PL measurements when excited by the 1.959 eV laser. The sample temperature was controlled by a programmable hot-stage THMS600 from Linkam Scientific Instruments Ltd.

Figure 1 shows the PL spectra of GaAs$_{1-x}$N$_x$ with $x =0.62\%$ by different excitations at 80 K. The PL peaks at 1.374, 1.721, and 1.860 eV are assigned as the optical transitions, $E_0$, $E_0 + \Delta_0$ (the spin-orbit split-off valence band), and $E_s$, respectively. The linewidth (full width at half maximum intensity) of $E_0$ is about 25 meV, and its peak energy and linewidth are found to be independent of the excitation energy. In contrast, the $E_s$ peak shows a surprisingly large linewidth, and both the peak position and linewidth vary significantly with excitation energy: a 13 meV blueshift in the peak energy (as indicated by two dashed lines in Fig. 1) and a change in the linewidth from 68 to 84 meV when excitation energy increases from 1.959 to 2.541 eV. The larger linewidth of $E_0$ in GaNAs alloys relative to that in GaAs is mostly from the usual inhomogeneous broadening, meaning that the energy variation of a specific state with the fluctuation in the local atomic configuration. However, the width of the $E_s$ band should have two contributions: the inhomogeneous broadening, which should be in the same order of magnitude of the $E_0$, and the energy spread of different

---

$^a$Electronic mail: pt290@cam.ac.uk and pinghengtan@hotmail.com
$^b$Electronic mail: yong.zhang@nrel.gov
perturbed k states, meaning that even N atoms are distributed in an ordered manner, there will still be an $E_0$ band (rather than a single state), which explains why the linewidth of $E_0$ is much larger than that of $E_0$. It is interesting to contrast the broadening of the $E_0$ band in GaAs:N with an extensively studied problem, the broadening of the N bound state in GaP$_{1-x}$As$_x$:N. For the latter, the alloy fluctuation of the $E_0$ bandwidth exhibits an abnormal temperature dependence, reflecting the unusual density of states and thermalization within the $E_0$ band. The shift of $E_0$ is found to be smaller than that of $E_0$, even though the temperature variation of the unperturbed host states at the $L$ point is larger than that at the $L$ point in GaAs, as indicated by the dashed lines in Fig. 2(b).

In a very similar sample, GaAsN alloy with 0.6% N, Francoeur et al. recently reported that the energy shifts of $E_0$ ($\sim$80 meV) and $E_0$ ($\sim$90 meV) between 80 and 300 K in MR measurements are fairly close to those of the band gaps at the $\Gamma$ ($\sim$85 meV) and $L$ (100 meV) point in GaAs. MR is a differential spectroscopy that amplifies the roles of few states near the “singularity” of the electronic density of states, and thus reflects the energies of critical points. PL, being a linear spectroscopy, may probe the contributions of states in a more broad range. If we assume approximately that the temperature change causes a rigid shift of the band structure in the MR measurement, the difference between the energy shift measured by MR and PL can then be attributed to the thermalization-induced blueshift. In general, the blueshift is determined by the density of states, the relative carrier relaxation time $\gamma = \tau_{\text{rel}} / \tau_{\text{rad}}$ ($\tau_{\text{rel}}$ is the relaxation time and $\tau_{\text{rad}}$ the radiative decay time), and excitation density. Under the assumption of parabolic dispersion and thermal equilibrium, the thermalization of free carriers will result in a blueshift of $k_BT/2 = 0.086T$ meV/K for the PL peak position relative to the band gap energy, where $k_B$ is Boltzmann’s constant. The temperature variation from 80 to 300 K will yield a blueshift of $\sim$10 meV. However, with respect to the critical energies determined by MR, the blueshifts are found to be $\sim$21 meV ($\sim 1.1k_BT$) and $\sim$42 meV ($\sim 2.2k_BT$), respectively, for $E_0$ and $E_0$, indicating an unusual behavior of...
carrier thermalization in the studied GaAsN alloy.

To quantitatively evaluate the behavior of carrier thermalization in GaAsN alloy, we extract the unusual thermalization-induced blueshifts in the GaAsN alloy, by taking the energy variations of the critical-point energies at Γ and L in GaAs as approximations for those of E₀ and Eₚ. If the blueshift is assumed to be a linear function of T, the PL peak energy in GaAsN can be fitted by the Bose-Einstein expression with an additional term for the thermalization-induced blue shift,

\[ E(T) = E_T - a_B(1 + 2/(e^{\Theta T} - 1)) + a_{CT} T, \]

where Θ represents the average phonon temperature, a_B the electron-phonon interaction strength, and a_CT the temperature coefficient for the carrier thermalization. Using the a_B and Θ parameters of GaAs at Γ and L points, we can fit the experimental data very well with Eq. (1) and obtain a_CT = 0.11, 0.06, and 0.21 meV/K for E₀, E₀ + Δ₀, and Eₚ in the GaAsN alloy. The results are summarized in Table I.

The parameter E_B in Table I shows the significant band gap change in GaAsN alloy with small amount of N incorporation into GaAs, compared with the fitting values for the critical-point energies of E Priest (1.570 eV), E₁ + Δ₀ (1.910 eV), and E₂ (1.872 eV) in GaAs. The fitting parameters a_CT yield the thermalization-induced blueshifts of 24 and 46 meV between 80 and 300 K for the E₀ and Eₚ transitions in the GaAsN alloy, respectively, which is much larger than that (10 meV) of k_BT/2. On the other hand, the corresponding blueshift (13 meV) of E₀ + Δ₀ is close to k_BT/2. We note that E₀ and E₀ + Δ₀ are associated with the common conduction band states, which indicates that the larger blueshift for E₀ is mainly resulted from the thermalization of the holes near the top of the valence band, probably due to the fact that the carriers are not in thermal equilibrium as it often occurs under the high excitation density, and thus the carrier temperature could be different from the lattice temperature. The major difference between Eₚ and E₀, which involves the same valence band states, should be attributed to the difference of the carrier thermalization for the conduction band states in the Eₚ band and near the band edge. The difference is highly anticipated because the density of states and the relative carrier relaxation times are expected to be very different near Eₚ and E₀. The density of states near E₀ is likely to remain as a monotonic function of energy, whereas the density of the perturbed states of the Eₚ band is not expected to be monotonic function of energy but with a limited extension.

Because of the unusual aspects of Eₚ, in its components and density of states and carrier relaxation, the Eₚ line shape and the thermalization effect are also expected to depend on excitation energy and power. As revealed in Fig. 1, different numbers of perturbed states are excited with different excitation energies, and thus the Eₚ line shape varies. These unusual aspects are also reflected in its excitation powder dependence, with a smaller exponent compared to E₀ or E₀ + Δ₀, as shown in the inset of Fig. 1. The smaller exponent of the Eₚ band could be understood as the result of more photoexcited electrons being populated to higher energy states, as evidenced by the line shape change, which is, in fact, consistent with the larger thermalization-induced blueshift. Finally, the slow down of the bandwidth increase for Eₚ above 240 K may indicate the finite bandwidth of the perturbed states because the states at the high-energy side are expected to be less and less perturbed.

In summary, we have reported a detailed photoluminescence study of the E₀ and Eₚ bands in a dilute GaAs₁₋ₓNₓ alloy with x = 0.62% using micro-PL technique, with varying temperature, excitation energy, and density. The temperature variations of the peak energies of E₀ and Eₚ in 0.62% GaAsN alloy have been found to be reduced, respectively, by 24 and 46 meV compared to those of the E₁ and Eₚ in bulk GaAs. The thermalization-induced unusually large blueshift suggests that the Eₚ band exhibits a unique density of states and carrier dynamics, which are very different from those of unperturbed host states in GaAs. The variations in the peak intensity and line shape of the Eₚ band with excitation energy and/or power, and the abnormal variation of linewidth with temperature can all be explained consistently.

This work was supported by NSF of China under Contract Nos. 60521001 and 10334040, the special funds for Major State Basic Research Project of China (G001CB3095) and NSF of Jiangsu (Grant No. BK2004403), the Grant No. HKUST 6076/02P by RGC of Hong Kong, China, and the U.S. DOE under Contract No. DE-AC36-99GO10337 at NREL.

<table>
<thead>
<tr>
<th>E₀ (eV)</th>
<th>a_B (eV)</th>
<th>Θ (K)</th>
<th>a_CT (10⁻³ eV/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.427</td>
<td>0.058</td>
<td>249</td>
<td>0.11</td>
</tr>
<tr>
<td>E₀ + Δ₀</td>
<td>1.785</td>
<td>0.062</td>
<td>249</td>
</tr>
<tr>
<td>Eₚ</td>
<td>1.913</td>
<td>0.065</td>
<td>249</td>
</tr>
</tbody>
</table>