EXCITON TRANSFER IN GaASxP1-x : N

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Résumé - Les mécanismes de transfert à basse température entre les états localisés sont étudiés par l'excitation sélective à l'intérieur de la bande non uniformément élargie des excitons liés aux impuretés azote dans GaAs$_x$P$_{1-x}$:N. Le mécanisme de transfert dominant est celui d'un tunneling par phonons acoustiques. Le coefficient de transfert est bien plus rapide que le coefficient de recombinaison radiative. Les formes des raies observées par excitation sélective, peut s'expliquer par la dépendance en énergie de l'interaction entre exciton et phonons, et par l'existence d'un "état final" d'où l'exciton ne peut effectuer un effet tunnel. Ces états sont directement observés par l'émission depuis la ligne spectrale la plus basse B' (J=2). Cette interprétation est confirmée par la levée de dégénérescence Zeeman quand un champ magnétique est appliqué.

Abstract - Selective excitation within the inhomogeneously broadened band of excitons bound to nitrogen impurities in GaAs$_x$P$_{1-x}$:N is used to study the low temperature transfer between localized states. The dominant transfer mechanism is by acoustic phonon-assisted tunneling. The transfer rate is much faster than the radiative recombination rate. The lineshapes observed under selective excitation can be explained by the energy dependence of the exciton-phonon interaction and the existence of "terminal states" from which the exciton cannot tunnel. These states are directly observed via the emission from the lower B'-line (J=2). This interpretation is further confirmed by its Zeeman splitting in an applied magnetic field.

I - INTRODUCTION

The study of exciton states in mixed III-V semiconductors was directed mainly at excitons bound to impurities. Of particular interest is the system of excitons bound to nitrogen impurities in GaAs$_x$P$_{1-x}$ and Ga$_x$In$_{1-x}$P /1-3/. The emerging physical picture for GaAs$_x$P$_{1-x}$:N is as follows: The band of excitons bound to nitrogen (N$_x$) is inhomogeneously broadened by the random potential fluctuations which arise from the distribution of microscopic environments of the binding N centers. Exciton transfer takes place between these nitrogen sites. Kash /4/ has shown that the N$_x$ luminescence lineshape under above-gap cw excitation can be interpreted as due to exciton recombination at "terminal states", namely, nitrogen sites from which no transfer can take place. A similar idea has previously been proposed to account for the undulation spectral shape in GaP doubly doped with N and Zn acceptors /5/. Time resolved spectroscopy /6,7/ indicated that transfer times within the N$_x$ band range around 1 nsec. In this work we provide a spectroscopic proof to the existence of terminal states within the N$_x$ band and show that the transfer process between nitrogen sites is determined mainly by the exciton interaction with long wavelength acoustic phonons.

II - EXPERIMENT

The crystals used in this study were bulk $GaAs_xP_{1-x}$ (x<0.35), lightly doped with nitrogen. They were grown from the vapor phase and had the form of thin needles or parallelepipeds with well developed faces. Luminescence spectra were excited with a dye laser (line width of ~0.12 Å) which operated either cw or pulsed, using a cavity dumper (pulse width of 7 nsec). The absorption and emission spectra were measured with a double monochromator having a spectral resolution of ~0.05 Å. All spectra reported here were taken with the samples immersed in liquid helium.
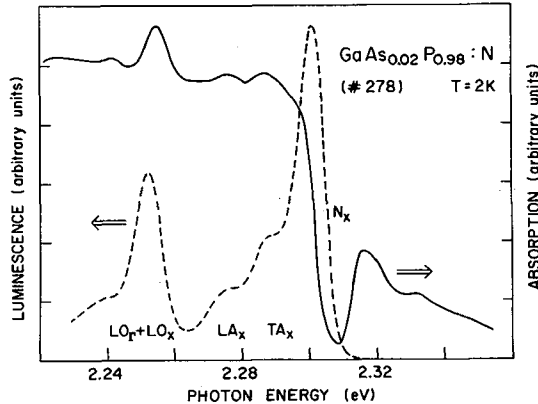

Fig. 1 - Absorption and luminescence spectra of a nitrogen-doped $GaAs_{0.02} P_{0.98}$ sample. Excitation is at 2.35 eV.

Fig. 1 shows the absorption and luminescence spectra of a $GaAs_{0.02} P_{0.98}$:N sample. The absorption spectrum shows the N_x band riding on a smeared band tail. The luminescence spectrum consists of the N_x band and its phonon sidebands. Note that the emission phonon sidebands are also seen in the absorption spectrum. Fig. 2 shows several emission spectra obtained under selective excitation within the N_x band. The nitrogen concentration in sample #278 is about 100 times higher than that in sample #549 (which was undoped) and contained only background impurities. The energies of the exciting laser lines are marked in the figures. In Fig. 3, high resolution luminescence spectra of the region just below the exciting laser lines, are shown. These were taken for nitrogen-doped samples with x = 0.98 and x = 0.65. The splitting of the sharp B'-line under an applied magnetic field of 28 kG is shown in Figs. 3a and 3c. Similar results were observed for several samples with $0.65 \leqslant x \leqslant 0.99$.

III - DISCUSSION

The luminescence spectra which are selectively excited within the N_x band consist of several bands: a relatively narrow band (denoted FF in Fig. 2) peaking 1.6 - 3.1 meV below the laser line and acoustic and optic phonon sidebands. The FF peak position, width and intensity, depend on the excitation energy within the band. There is also a marked dependence on the nitrogen concentration (cf. Figs. 2a and 2c): the FF band is much narrower for the sample with low nitrogen content. Fig. 4 shows the integrated intensity of this band as a function of excitation energy for $GaAs_{0.02} P_{0.98}$:N. Also shown is the luminescence spectrum obtained under above-gap excitation. We interpret this band as due to the recombination of
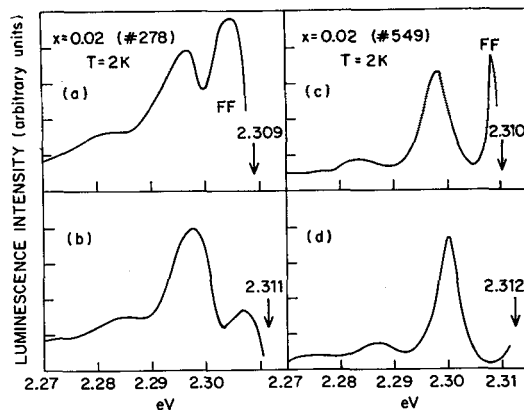
Fig. 2 - Selectively-excited luminescence of a nitrogen-doped (a,b) and undoped (c,d) $GaAs_{0.02} P_{0.98}$ samples. Arrows mark the exciting laser energies.
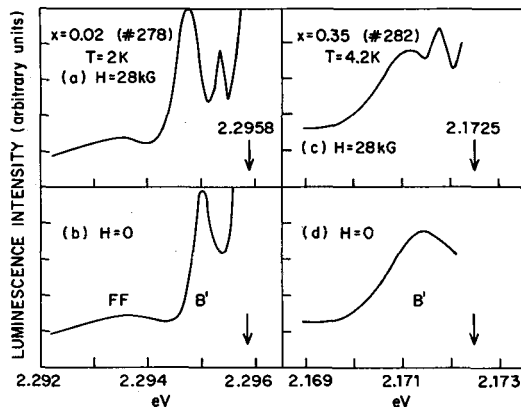

Fig. 3 - Sharp line structure of the selectively excited luminescence spectrum just below the laser line for two compositions of $GaAs_x P_{1-x}$. The Zeeman splitting is shown in a and c.

excitons redistributed between localized states by acoustic phonon-assisted tunneling /8/. For the case of piezo-electric coupling between the exciton and acoustic phonons, the interaction form factor has a maximum strength for an energy difference of $\Delta E = 1.6$ meV between the initial and final exciton states. This matches the observed peak position of the FF band in sample #549 which contains only background N impurities. This means that for low nitrogen content, only single transfer steps take place. For sample #278 which has about 100 times more N impurities the FF band peak shifts to 3 meV and the width increases. This indicates that multiple transfers occur. Collet et al. /6/ measured transfer times of the order of 1 nsec. Our time resolved spectra show that within the resolution of the set-up (7 nsec), the

observed lineshapes are the same as those taken with cw excitation (Fig. 2). The
radiative lifetime (measured at several points along the N_x band) is about 300 nsec
for x = 0.98. Therefore, it is clear that the FF band shape is not determined by
a simple competition between the radiative recombination and exciton transfer with-
in the N_x band. This supports the idea of terminal exciton states /4,5/: at T=0,
transfer processes take place between localized states until the exciton reaches a
nitrogen site which is not connected with any other lower-energy site. The exciton
is thus trapped on the terminal state and radiatively recombines there.
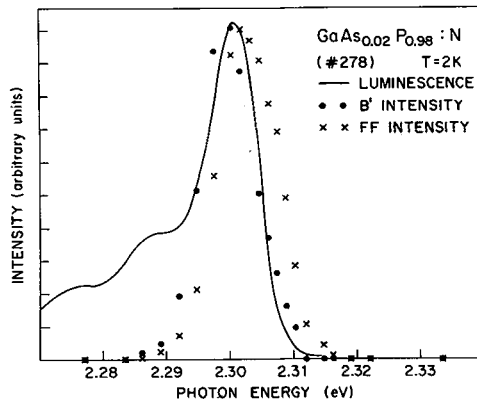

Fig. 4 - The integrated luminescence intensity of the FF band   , B' line   and
the luminescence spectrum excited above the gap. (All normalize to unity at peak).

We have directly measured the relative density of terminal states within the N_x band
by utilizing the splitting of the exciton 1S state into J=1 and J=2 states which
are separated by about 0.8 meV. These states give rise to the A' and B' lines in
analogy to GaP:N /9/. Selective excitation into the upper J=1 state of a given
bound exciton is followed by rapid thermalization into the lower J=2 state (of the
same exciton). Emission then occurs from the J=2 state. Examples
of high resolution spectra which allow the observation of the B' line are shown in
Fig. 3b and 3d. The assignment is verified by observing the Zeeman splitting of the
B' line, which, for x = 0.98, is identical to that of the B' line in GaP:N /9/. The
relative intensity of the B' line as a function of excitation energy within the N_x
band is shown in Fig. 4, and it gives the relative density of terminal states.

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