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STUDY ON THE LUMINESCENCE OF GaP:N UNDER SELECTIVE EXCITATION OF EXCITONS BOUND TO NN_1 CENTERS*

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Under selective excitation of excitons bound to NN_1 centers, we observed and studied the luminescence of excitons bound to shallower NN_i (i=3, 4, 5), isolated nitrogen centers and of free excitons.

1. INTRODUCTION

The photoluminescence of excitons bound to isoelectronic traps in GaP:N has been studied for many years. Most of the properties of various NN_i centers are well known now[1]. To our knowledge, in all the previous works the excitation photon energies are higher than the emission photon energies.

As there exist nonradiative decay, Auger effect and tunneling of the whole exciton to another centers other than the radiative process[2], the possibility of deeper bound excitons turning to shallower bound exciton is reasonable. And if the excitation density is high enough, the phenomenon should be observable[3].

At low temperature, under selective excitation of excitons bound to NN_1 centers, we observed the luminescence of excitons bound to NN_i (i=3, 4, 5), isolated nitrogen and of free excitons at the high energy side of exciting laser. We propose that this phenomenon is due to the tunneling effect of bound excitons with the assistance of phonon annihilation or Auger effect. We have also done the luminescence dynamics analysis on the experimental results.

2. EXPERIMENT

We have used a Nd:YAG pumped tunable dye laser whose wavenumber range is from 17640 to 18050 cm^{-1}. The dye laser power density can reach as high as $3x10^7$ W/cm^2 at the focus point of lens. The pulse temporal width is about 7 ns and the pulse repetition frequency is 15 Hz. The GaP:N sample was grown by liquid phase epitaxy method whose nitrogen concentration was about $6x10^{17}$ cm^{-3}. The sample was placed in a cryogenic system, the sample temperature can be changed from 8 K to 300 K. The emission light of sample was collected into a SPEX double grating monochromator, detected by a photomultiplier and through a Boxcar averager the signal was inputed to digital microprocessor.

At 8 K, tuning the wavenumber of dye laser to 17857 cm^{-1} (about the energy of an exciton bound to NN_1 plus the energy of a LA phonon of GaP:N), under high density excitation, we have measured the luminescence of excitons bound to NN_1 and its phonon replica and that of excitons bound to NN_3, NN_4, NN_5, isolated nitrogen and of free excitons, as shown in the figure. The position of FE is 18784 cm^{-1}. Its energy is about 12 meV higher than that of excitons bound to isolated nitrogen.

We measured the luminescence intensity as a function of sample temperature. The temperature was changed from 10 K to 100 K. Because the band gap of GaP:N will change as sample temperature is raised, we slightly changed the exciting laser wavenumber for different temperature to match the variation of band gap. The excitation

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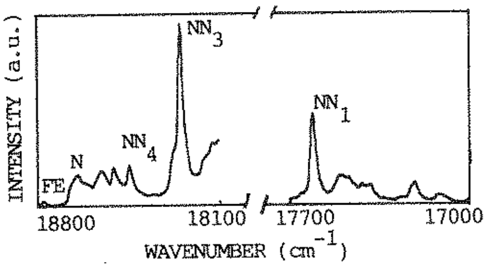

FIGURE
Luminescence spectra of GaP:N. The excitation wavenumber is 17857 cm^{-1} and excitation density is $2x10^7$ W/cm^2

density was $2x10^7$ W/cm^2. The luminescence of NN_4, NN_5 decreased as the temperature rose, similar to their behaviours under band gap excitation condition. From 20 K to 100 K, the luminescence of NN_3 decreased. The luminescence intensity of NN_1 changed only a little and the FWHM increased obviously from 10 K to 100 K.

At 8 K, from $2.4x10^5$ W/cm^2 we increased the excitation density about 100 times gradually to measure the intensity of the luminescence of both NN_1 and NN_3 centers. From $2.4x10^5$ to $8x10^5$ W/cm^2, there was only the luminescence of NN_1 and its phonon replica increasing with the excitation density. From $8x10^5$ to $4x10^6$ W/cm^2, the luminescence of NN_3 appeared and increased faster than that of NN_1 did, but the intensity of NN_3 was weaker than that of NN_1. From $4x10^6$ to $2x10^7$ W/cm^2, the luminescence of NN_3 increased faster and its intensity became stronger than the intensity of NN_1, as shown in the figure for the excitation density of $2x10^7$ W/cm^2.

3. ANALYSIS

The experimental results suggest that the tunneling effect of the whole exciton bound to NN_1 can be very strong under high density excitation condition. While the selective excitation density is so high that the density of excitons bound to NN_1 is close to the concentration of NN_1 centers, the interaction between bound excitons and lattice and between bound excitons become very strong. Maybe these make the exciton tunneling effect obvious.

The dependence of the intensity of the luminescence of NN_1 on temperature in our experiment was different from that under weak excitation condition. This difference is easy to be understood considering that the time spent to reach thermal equilibrium of excitons population and the time of tunneling process are much shorter than the time every excitation pulse lasts (7 ns) while the excitation density was very high.

Because the number of NN_3 pairs is larger than that of NN_1 and the tunneling effect is strong under high density excitation, the population of excitons bound to NN_3 can be larger than that of excitons bound to NN_1. So the luminescence of NN_3 will be stronger and increase faster than that of NN_1 from a particular excitation density. The particular density may be the density that saturates the NN_1 centers.

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