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THE PHONON SIDEBANDS OF NN_i PAIR EMISSION IN GaP:N

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By using selective excitation technique, the phonon sidebands belonged to each NN_i pair center were successfully resolved out from the seriously overlapping spectrum under above gap excitation. The TA and LA phonon sidebands of NN_3 center were clearly seen at low temperature. This work confirmed that NN_1 ,NN_3 and isolated N centers have almost the same phonon sideband structure.

1. Introduction

The luminescence of isoeletronic impurity bound excitons in GaP:N have been investigated quite extensively and intensively (1-8). The coupling of the bound excitons with phonons is one of the most interested problems in this area. Not only for the importance on this problem itself, the study on it also connected with the study on the binding mechanism of isoelectronic traps (4-8). Owing to the fact that the potential of the isoelectronic trap is strongly localized, the phonon sidebands of the bound exciton recombination basically reflect the density of phonon states. The emission lines of the excitons bound to isolated N center or NN_i pair centers show almost the same phonon structure (1,8). They are all composed of TA, LA, TO^{Γ}, LO^{Γ} and "X" phonon sidebands ("X" phonon might be attributed to the minimum of LO branch in the K-point (9)). The little difference is that NN_i pair centers still show their weak local-mode phonon sidebands, but isolated N center does not. Such a similarity is very obvious when one compare NN_1 center to isolated N center. Since the level of NN_1 center is quite far from the other NN_i centers, there is almost no overlapping between their emission lines. However, when one compare the other NN_i centers to isolated N center, the similarity is ambiguous because the overlapping of the phonon sidebands from diffrent centers make it difficult to distinguish them from each other in the spectrum. By carefully analysising the temperature-variation photoluminescence spectra, Ref.(8) found as T > 30K NN_3 center showed the same phonon sideband structure as that of isolated N center or NN_1 center, that is, NN_3 center showed apparent TA and LA phonon sidebands. Neverthless, at lower temperature, the emission of NN_3 center was relatively weak, so its TA and LA phonon sidebands were submerged by the LO^{Γ} and "X" phonon sidebands of NN_4 ,NN_5 and NN_6 centers. For the other NN_i (i > 3) centers, their TA and LA phonon sidebands are more difficult to be observed.

In this work, by selectively exciting with the energies of corresponding to NN_3 , NN_3 +TA^x , NN_4 and NN_5 lines, we have confirmed that at low temperature the phonon sideband structure of NN_3 center is just the same as that observed in Ref.(8) at higher temperature, verifying that NN_1 , NN_3 and isolated N centers have the same phonon sidebsnd structure.

2. Experimental Results and Analysis

The sample was epitaxially N-dopped GaP crystal, nitrogen concentration was about 2.4×10^{18} cm^{-3} (8). In the experiment, the sample was cooled to 15K in a DE-202E cryostat, and the temperature was adjustable.

The luminescence was excited by a Model 375 tunable dye laser pumped with a Spectra-Physics 171 Ar-ionized laser. The dye used was Coumarin 6. The luminescence was detected by a system including Spex 1403 monochrometer, cooled RCA C31034A photomultiplier, and PAR 5206 lock-in amplifier.

Fig.1 is a photoluminescence spectrum of GaP:N at low temperature with above gap excitation (8). One can see in Fig.1 that the region 250 cm^{-1} below the zero-phonon line of NN_3 center is rather crowded, many phonon sidebands from different NN_i centers lie closely in this region. At low temperature, as the luminecent intensities of NN_4, NN_5 and NN_6 centers are relatively strong, their phonon sidebands dominate the region and so the TA and LA phonon sidebands of NN_3 center can not be seen obviously. But, by selective excitation, one can clearly distinguish the contribution from each NN_i pair center in the spectrum. Fig.2 shows a set of spectra by selectively exciting with energies corresponding to $NN_3 + TA^X$, NN_4 and NN_5 lines respectively at 15K. In Fig.2(a), excited with the energy of $NN_3 + TA^X$, only those phonon sidebands related to NN_3 center appear in the above mentioned region. Since the excitation energy is lower than the levels of other NN_i (i > 3) centers, the phonon sidebands of these centers should not appear in this case. Also, the same result was obtained when excited with the energy of NN_3 line. In Fig.2(b), excited with the energy of NN_4 line, NN_4^* emerges at higher side of NN_3-LA band (here NN_i^* standing for two phonon sidebands NN_i-LO^Γ and NN_i-X), and NN_3 -LA band is now almost unobservable. Because of resonant excitation to NN_4 level, NN_4^* emission is quite strong, however, the emission of NN_3 center is rather weak since it is only induced by the $NN_4 \rightarrow NN_3$ excitonic tunneling transfer (10). In Fig.2(c), excited with the energy of NN_5 line, another new band NN_5^* appears at the lower side of NN_3-TA band. Also, for the existence of $NN_5 \rightarrow NN_4$ and $NN_5 \rightarrow NN_3$ excitonic tunneling transfer, the emission lines of NN_4 and NN_3 centers appear in the same time. In this case, the spectrum shows a complex structure as in Fig.1.

Fig.3 is the luminescence spectra at 54K. When excited with the energy of $NN_3 + TA^X$, the spectrum still has only the phonon sidebands of NN_3 center, as shown in Fig.3(a). When excited with the energy of NN_4 line, the NN_4^* is added to the spectrum. But, for the thermal dissociation of exciton bound to NN_4 center, the intensity of NN_4^* decreased obviously, so NN_3 -LA band becomes observable in the spectrum. In fact, with the above gap excitation, if T > 50K, the intensity of NN_4^* was much weaker than that of NN_3 -LA (8). At present, because of resonant excitation to the level of NN_4 center, NN_4^* still

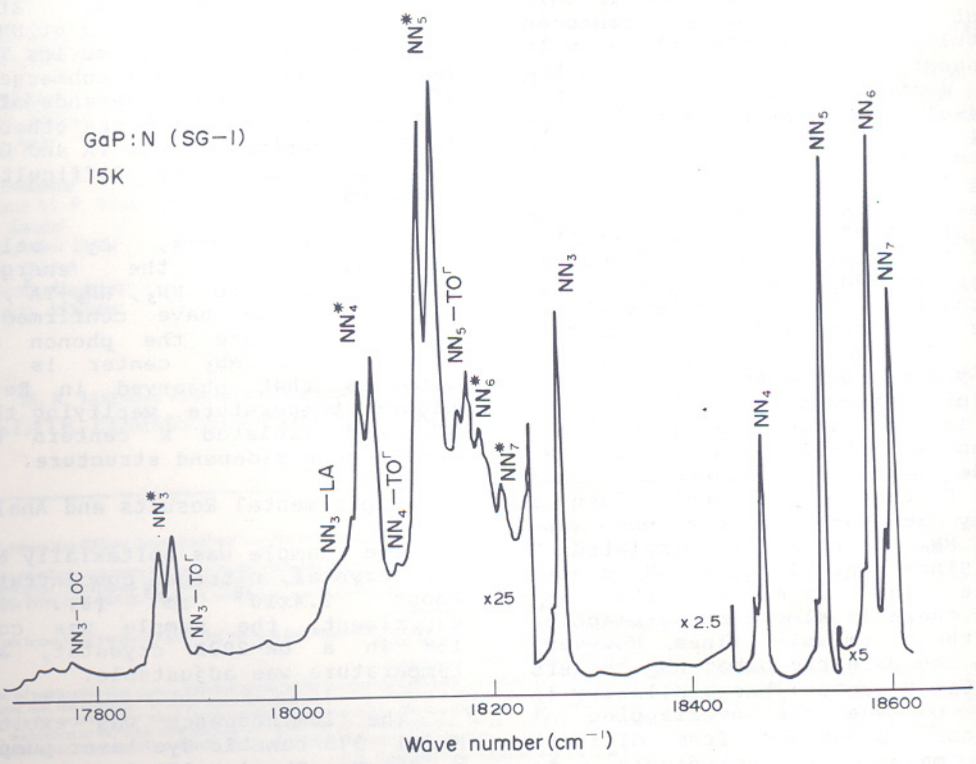

Figure 1.

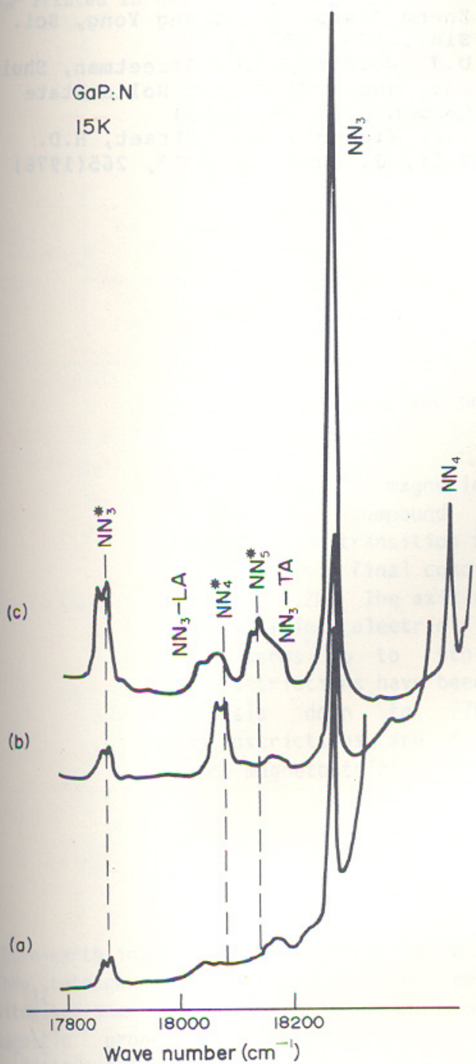

Figure 2.

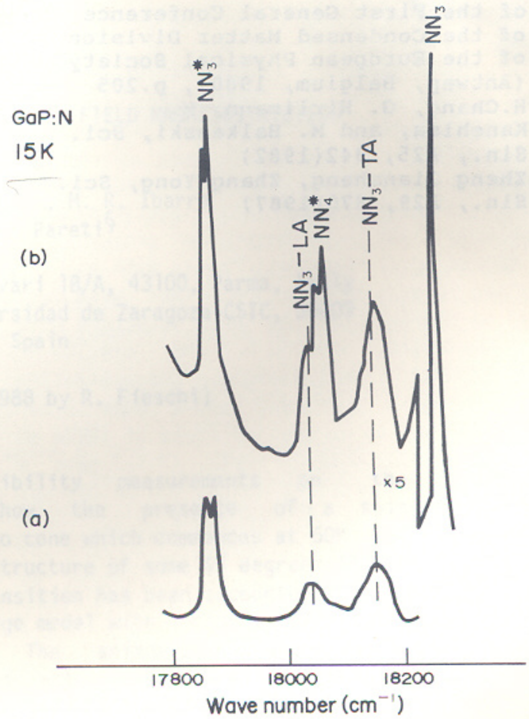

Figure 3.

shows fair intensity. Also, the luminescence of NN_3 center increases as two kinds of energy transfer processes, tunneling transfer and thermal ionization transfer, take place at the same time (7).

3.Conclusions

By using selective excitation technique, the phonon sidebands belonged each NN_i pair center were successfully resolved out from the seriously overlapping spectrum under above gap excitation. Thus, this work has confirmed the assignment to the TA and LA phonon sidebands of NN_3 center in Ref.(8), and so make it clear that the anomalous enhancement (5,6) of LO phonon sidebands of NN_4 , NN_5 and NN_6 centers at about 50K was not due to the temperature variation of the Huang-Rhys' factors, but due to the mixing with TA and LA phonon sidebands of NN_3 center. This study shows that both NN_1 and NN_3 cenetrs have rather strong LO^{Γ} , "X", TA and LA phonon sidebands as well as isolated N center, which indicates that the bound excitons have a fairly strong coupling with these phonons. This study provides a strong support to the conclusion made in Ref.(8): that the coupling of nitrogen-bound excitons with phonons in GaP:N accords with the Huang-Rhys' multiphonon optical transition theory.

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