

Deep-center photoluminescence in nitrogen-doped ZnSe

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In this paper, we report optical studies of nitrogen-doped ZnSe epilayers grown by molecular-beam epitaxy. Photoluminescence spectra of the donor-acceptor pair region at different temperatures and different carrier concentrations show that two donors are present in the samples: residual shallow donors with activation energy 26 meV and deep donors with activation energy of 4 meV previously assigned to a V_{Se} -Zn- N_{Se} complex. In the exciton region, we observe an emission at 2.765 eV, which shows an increased intensity when the epilayer is compensated by the deep donor. We therefore propose that this transition may be related to a deep-donor bound exciton.

I. INTRODUCTION

There has been a long history of attempts to produce *p*-type ZnSe. Park *et al.*¹ and Ohkawa *et al.*² successfully doped ZnSe grown by molecular-beam epitaxy (MBE) using nitrogen as a dopant, and this important step led to the demonstration of the II-VI blue-green semiconductor laser diode.³⁻⁵ Qiu *et al.*⁶ examined the photoluminescence spectra over a range of doping levels and showed that ZnSe:N is characterized by intense donor-acceptor pair (DAP) emissions. They reported *p*-type doping levels up to $(N_A - N_D) = 10^{18} \text{ cm}^{-3}$, but additional nitrogen is fully compensated and so we must understand the role of nitrogen in the ZnSe in order to increase the number of active nitrogen acceptors.

Nitrogen forms an acceptor in ZnSe with an activation energy of 110 meV.⁷ Detailed photoluminescence (PL) measurements by Hauksson *et al.*⁸ showed that a DAP emission occurred in highly nitrogen-doped ZnSe and that a deep compensating donor with a binding energy of approximately 44 meV existed in this material. It was proposed that the donor is a complex consisting of a nitrogen acceptor and a doubly charged selenium vacancy donor on a next-nearest-neighbor site (V_{Se} -Zn- N_{Se}). This complex would therefore be a single donor. The emission due to transitions from deep donors to free holes (D^0F) was observed by Zhu *et al.*,⁹ which suggested a deep donor binding energy of 55 meV.

Recent optically detected magnetic resonance (ODMR) results¹⁰ on the same samples showing both shallow and deep DAP luminescence showed signals due to the shallow isotropic donors and deep anisotropic donors, consistent with the model proposed in Ref. 8. Three ODMR resonances were observed having $g = 1.11$ for the 26-meV shallow donors, $g = 1.38$ for the 44-meV deep donor, and $g = 2.00$, which was initially assigned to the 110-meV deep nitrogen acceptor but more recently shown to have anisotropy consistent with a $\langle 111 \rangle$ center such as (V_{Se} -Ag) or (V_{Se} -Au) complexes.¹¹ Recent spin-flip Raman

spectroscopy agrees with these assignments.¹² ZnSe:N samples were measured by capacitance-voltage (C - V) profiling four months after growth and changes in the net active dopant concentration were observed.⁸ Similar changes were also seen in photoluminescence spectra, where the deep DAP transitions increased relatively to the shallow DAP over a period of six months. These results were interpreted to suggest that vacancies diffuse from the layer surface and the creation of deep donor therefore continues after growth at room temperatures. Calculations of vacancy concentrations as a function of temperature and selenium and zinc overpressures have shown that the material is always undersaturated with vacancies and that there is a substantial concentration gradient, leading to the subsequent indiffusion of vacancies.¹³

In this paper we examine the radiative recombination processes in nitrogen-doped ZnSe grown by MBE and report detailed measurements of the temperature dependence of the emission for different nitrogen doping levels, which show that the shallow and deep donors are associated with the nitrogen acceptors in the DAP recombination. Excitation power-dependent measurements show the presence of deep impurity transitions, possibly associated with deep donor-acceptor pairs.

II. EXPERIMENTAL DETAILS

The epilayers were grown by molecular-beam epitaxy using a Vacuum Generators VG288 growth system and details are given elsewhere.¹⁴ For *p*-type doping we used a nitrogen rf plasma source supplied by Oxford Applied Research. For some samples, part of the layer was illuminated by above band-gap radiation during growth using a krypton ion laser (351 nm) with intensities up to 4 W cm^{-2} . The incorporation of active nitrogen is affected by the illumination and it has been shown that doping levels are increased.¹⁵ For the samples investigated in the present study, a diameter of approximately 5 mm was

illuminated through a heated quartz window. Thus a range of doping levels can be explored by observing the PL from different parts of the crystal. For example, the maximum nitrogen concentration will be at the center of the illuminated region and the minimum outside the laser spot. However, because of compensation the maximum ($N_A - N_D$), where N_D is the donor concentration and N_A the acceptor concentration, is not necessarily at the spot center.

Measurements were made on samples of various carrier concentrations. The PL data presented here are from six samples, Nos. 1–6, which are all ZnSe layers of approximately 1.5 μm thickness on (100) GaAs substrates. Sample No. 6 was not illuminated during growth. The uncompensated acceptor concentration ($N_A - N_D$) was measured by electrochemical C - V profiling¹⁶ and was found to be uniform in the direction of growth. Doping levels are given in Table I for each sample.

The PL measurements were performed using either excitation from a cw Ar^+ ion gas laser at 351 nm or from a cw Ar^+ -pumped blue laser using Stilbene 3 dye. The samples were immersed in liquid He for temperatures from 1.7 to 4.2 K and an Oxford Instruments cryostat was used to vary the temperature from 4 K up to room temperature. Dispersion of the luminescence was provided by a SPEX 0.85-m or an ISA 1.0-m double monochromator and the signal was detected with a cooled GaAs photomultiplier tube. In the measurements of the excitation intensity dependence of the emission, the excitation intensity was varied using neutral density filters, thus avoiding changes in the size or location of the laser spot on the sample.

III. RESULTS

A. Bound excitons

Figure 1 shows the PL spectra in the exciton region from three regions of a nitrogen-doped ZnSe at 4 K, where the different regions correspond to unirradiated, partly irradiated, and fully irradiated parts of the crystal and therefore correspond to different doping levels. Spectra for all regions are dominated by the acceptor-bound exciton (A^0, X) strain-split emissions at 2.7893 and 2.7912 eV, which is consistent with the fact that the sample is p type. At higher energies donor-bound and free-exciton features are also detected in each case; the identity of the latter was confirmed using reflectance. The strongest donor-related peak is that involving ionized donors (D^+, X), which is consistent with the expectation that all shallow donors should be ionized at equilibrium in p -type material. The (D^+, X) peak is similarly dominant over neutral donor-bound excitons in p -type

ZnSe:Li, as discussed by Zhang *et al.*,¹⁷ who use the notation of I_3 for (D^+, X). The emission at 2.758 eV is the LO phonon replica of the (A^0, X) doublet.

While the above peaks are all well known in nitrogen-doped material, we also observe two other features in the spectra of Fig. 1 at 2.765 and about 2.784 eV, respectively. The intensities of both peaks increase with nitrogen incorporation, as evident when comparing, e.g., spectrum (c) at the position of highest nitrogen doping (just inside the laser spot) with spectrum (a) at the point of lower doping (just outside the laser spot). Neither feature has been discussed before, as far as we know, although a peak similar to the one at 2.765 eV is evident in spectra published previously by Park *et al.*¹

We attribute the structure extending from the nitrogen (A^0, X) peak down to about 2.784 eV to excitons bound to pairs of neutral nitrogen acceptors (N-N) at various separation distances within the lattice. This type of structure is well known for a number of different acceptors, e.g., in ZnTe, where it is referred to as an undulation spectrum.^{18,19} A similar structure has been reported previously for copper²⁰ and phosphorus²¹ acceptors in ZnSe. The existence of this structure is due to the heavy acceptor doping in this material, since heavy doping is necessary to favor a significant concentration of close pairs, based on the Poisson statistics of the presumably random impurity distribution. The increased doping inside the laser spot is clearly responsible for the increase in the strength of the undulation wing relative to the (A^0, X) peak in Fig. 1(c) compared to Fig. 1(a). In agreement with previous observations,^{18,19} the undulation wing quenches rapidly in favor of the isolated nitrogen (A^0, X) doublet as the temperature is raised through the 10–30 K range (not shown). This effect is due to thermal excitation of the excitons from the close pairs to which they tend to tunnel at low temperature to the more common distant pairs and to essentially isolated centers.^{15,16} In some nitrogen-doped MBE ZnSe, the undulation wing actually dominates over the isolated (A^0, X) peak at low temperature; the latter is sometimes visible only as a weak shoulder on the lower energy peak, although it always becomes dominant as the temperature is raised.²²

The origin of the 2.765-eV peak is less clear. While it is nearly coincident (allowing for strain) with the reported position of the no-phonon line involving excitons bound to isoelectronic Te centers in $\text{ZnSe}_{1-x}\text{Te}_x$ with $x < 0.01$,^{22–24} it does not exhibit the pronounced phonon side bands that always accompany the Te-related emission (we verified this fact by a direct comparison to data we recorded on $\text{ZnSe}_{0.993}\text{Te}_{0.007}$ under the same conditions). Also, it is characteristically broader than the Te-related peak, and no Te had been used in the MBE growth system prior to the growth of these samples. For

TABLE I. Active nitrogen concentration (cm^{-3}).

$N_A - N_D$	Sample No.					
	1	2	3	4	5	6
Outside laser spot	1×10^{17}	4×10^{17}	2×10^{17}	7×10^{16}	3×10^{16}	
Inside laser spot	1.5×10^{17}	2.5×10^{17}	1.9×10^{17}	2.1×10^{17}	1.6×10^{17}	1×10^{17}

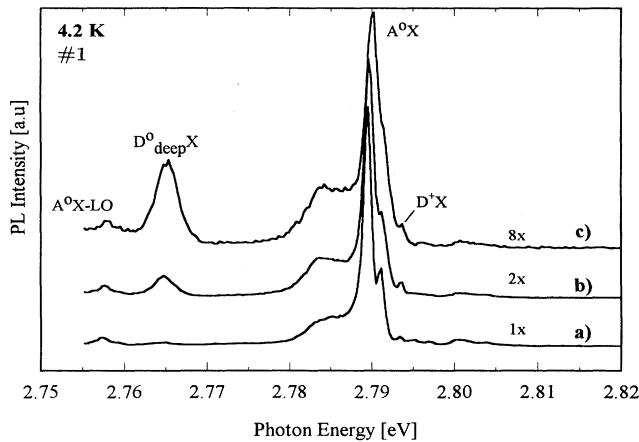


FIG. 1. PL spectra for sample No. 1 taken at 4 K. The three different spectra are taken at different positions, (a) just outside the laser spot, (b) on the edge of the laser spot, and (c) just inside the laser spot. Going from (a) to (c), $(N_A - N_D)$ changes from 1×10^{17} to $5 \times 10^{17} \text{ cm}^{-3}$.

these reasons, we rule out its assignment to Te.

The 2.765-eV peak has the following properties: (1) Its intensity relative to other bound exciton peaks increases with increasing N doping (see Fig. 1); in particular, it is strong in our samples that exhibit the deep DAP pair peak. It is frequently (Fig. 1 and Ref. 1) observed in heavily nitrogen-doped material, although it is very weak or absent in other heavily nitrogen-doped material that is nonetheless dominated by the deep DAP pair band.²² (2) It does not shift perceptibly with excitation intensity, ruling out any interpretation as a very shallow DAP band. (3) It quenches rapidly as the temperature is raised, disappearing by about 20 K. (4) It is characteristically broader than other bound exciton peaks such as the nitrogen (A^0, X) peaks in the same sample (its FWHM in Fig. 1 is about 3 meV). This property has been confirmed using a variety of samples from various sources. (5) Assuming that it is a bound exciton, the exciton localization energy deduced from the peak position is about 38 meV, compared to 10 meV for excitons bound to nitrogen acceptors. (6) The peak exhibits relatively weak coupling to LO phonons, which tends to favor its assignment to a center involving donors, rather than deep acceptors or isoelectronic centers. (7) It shifts to lower energy with biaxial tensile thermal strain at about the same rate as the nitrogen (A^0, X) peak (based on samples with differing thermal strains), showing that the shear deformation potential of the hole in the complex is not significantly quenched in comparison to hydrogenically bound holes. This observation suggests that the hole in the bound exciton is not highly localized (as does the phonon coupling strength, mentioned above).

Because of the intrinsic width of this peak, it is not possible to use perturbation spectroscopy to identify its origin more clearly. The width suggests a possible unresolved convolution of peaks involving several different centers with slightly different properties, such as different orientations with respect to the biaxial strain axis. However, no fine structure can be resolved. It is very tempt-

ing to attribute it to excitons bound to the new deep donors observed in the deep DAP band. However, the localization energy is surprisingly large compared to that of ordinary shallow (D^0, X) peaks (10 vs 38 meV), given that the donor binding energy increases only from ~26 to 46 meV. Also, the apparent absence of this peak in some samples exhibiting the deep DAP (Ref. 22) is difficult to understand if this interpretation is valid, although sample-dependent nonradiative recombination rates and the resulting positions of the electron quasi-Fermi level may play a role in the observation of the peak. In the absence of any clear alternative models, we tentatively assign it to excitons bound to the new deep neutral donors. This model explains most of the experimental properties listed above.

B. Donor-acceptor pair emission

Figure 2 shows PL spectra at various temperatures for a sample that shows only shallow DAP emission ($N_A - N_D = 7 \times 10^{16} \text{ cm}^{-3}$). The spectrum at 4 K shows dominant shallow DAP emission and its LO phonon replicas ($\hbar\omega_{LO} = 31.5 \text{ meV}$ for ZnSe). The free-to-acceptor ($F-A$) emission is observed as a shoulder on the high-energy side of the no-phonon DAP emission. The Y_0 transition is also observed, but this emission has been related to lattice defects.²⁵ As we increase the temperature the intensities of the DAP and its phonon replicas decrease as the donors ionize and the $F-A$ transition with its phonon replicas become dominant at 50 K. At 145 K the donor-to-free hole ($D-F$) emission has become dom-

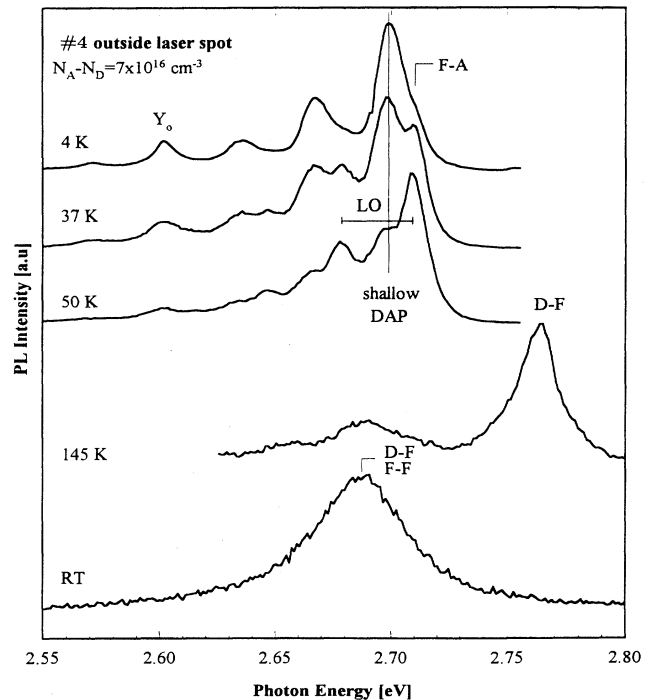


FIG. 2. PL spectra of sample No. 4 at different temperatures. The spectra are taken outside the lasers plot where $(N_A - N_D) = 7 \times 10^{16} \text{ cm}^{-3}$.

inant, indicating that the acceptors are partially ionized, while at room temperature the D - F or free-electron-to-free-hole (F - F) emission dominates.

Figure 3 shows the PL spectra for a sample with more active nitrogen ($N_A - N_D = 2 \times 10^{17} \text{ cm}^{-3}$), where we observe two distinct DAP transitions. The difference in the quenching behavior of the two different DAP transitions as the temperature is increased is strong evidence for the existence of two donors with different activation energies, as proposed by Hauksson *et al.*⁸ We clearly see that the shallow DAP emission decreases faster than the deeper DAP emission and this observation, combined with the detection of only a single F - A band, demonstrates that there are two donors with different activation energies involved. For temperatures higher than 40 K, the intensity of the deep DAP emission decreases very rapidly, and at 70 K the F - A and deeper transitions dominate the spectra. In fact, these deeper transitions dominate the emission from highly doped material but even at this doping level they are clearly observable. We will later show that these deeper transitions are DAP emissions, which dominate the spectra for carrier concentrations $> 2 \times 10^{17} \text{ cm}^{-3}$. At higher temperatures, the luminescence evolves towards band-to-band recombination.

Figure 4 shows the PL spectra at different temperatures for a sample more heavily doped than the one discussed above. Only deep DAP transitions at 4 K and the corresponding phonon replicas are observed. When the temperature is increased a free-to-acceptor transition appears, indicating that the 110-meV acceptor is still present. At higher temperatures (150 K up to room temperature) F - A and near-band-edge transitions dominate the spectrum.

Another observation regarding the DAP peaks that has not been previously emphasized is the strength of the LO phonon coupling for each set of peaks. By fitting the DAP spectra, we find values for the Huang-Rhys coupling factor S of 0.54 ± 0.04 for the shallow DAP peaks and 0.67 ± 0.04 for the deeper ones, respectively. The increase in S is qualitatively consistent with the increased localization of the electron bound to the deeper donor

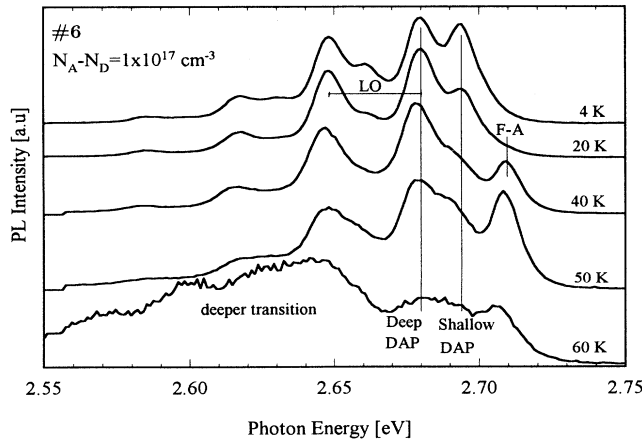


FIG. 3. Temperature-dependent PL of sample No. 6. Excitation intensity is approximately 0.1 W cm^{-2} .

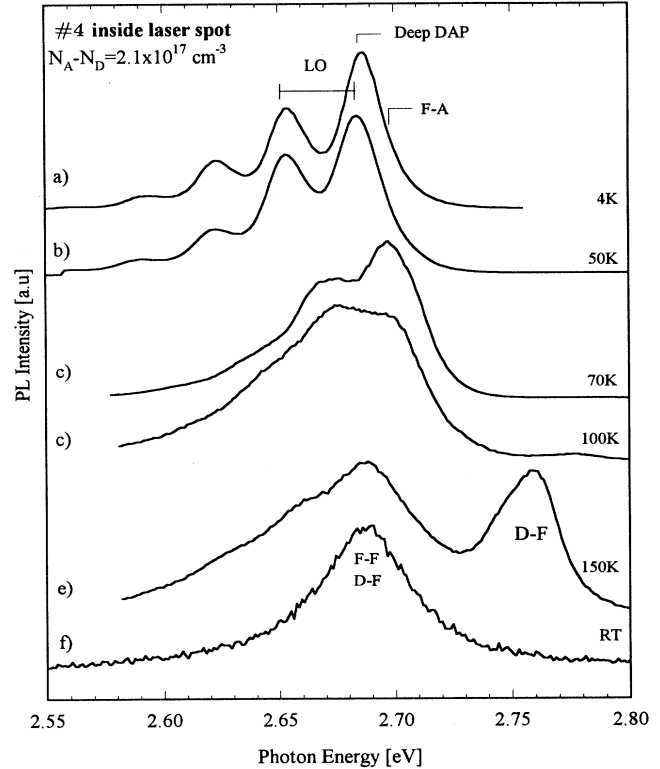


FIG. 4. Temperature-dependent PL of sample No. 4 taken inside the laser spot where the net doping level, $(N_A - N_D)$, is $2.1 \times 10^{17} \text{ cm}^{-3}$.

since increased localization in either the donor or acceptor wave functions increases S .²⁶

C. Selective pair luminescence

The variable temperature measurements discussed above display only a single F - A peak corresponding to the two observed DAP peaks, suggesting that the lower-energy DAP peak involves a deeper donor rather than a deeper acceptor level. However, this conclusion is subject to some uncertainty, because the F - A peak involving the deeper acceptor level (if one existed) would essentially overlap the shallower DAP peak, and might therefore not be clearly observable. To establish positively that a deeper donor is involved, we performed selectively excited donor-acceptor pair luminescence (SPL) measurements using a tunable dye laser. The principles of this technique have been given elsewhere.^{27,28}

Figure 5 displays a set of SPL spectra for the lightly doped sample No. 5, for six different energies of the exciting laser. The nonresonant DAP spectrum of this sample exhibits only the "normal," shallow DAP peak involving the ~ 110 -meV nitrogen acceptors. The spectra in this figure display sharp peaks involving either Raman satellites of the laser line or selectively excited DAP pairs, superimposed on broad, nonresonantly excited DAP peaks excited by virtue of tunneling of the excitation from the selectively excited close pairs to other pairs. The spectrum for the highest laser energy, which is resonant with

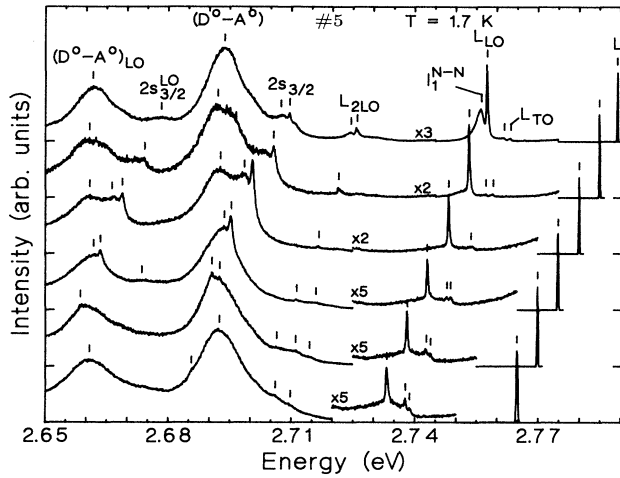


FIG. 5. Selectively excited DAP pair spectra for a lightly N-doped sample, recorded for six different laser energies. The peak denoted L (which has been greatly attenuated) is the laser line.

the (A^0, X) peak involving nitrogen, also exhibits a PL peak involving the excitons bound to N-N pairs and its phonon replicas. We focus here on the strongest SPL peak involving the $2S_{3/2}$ excited state of the shallow nitrogen acceptors, which is 79.3 meV below the laser line in the limit of distant pairs (low excitation energy). The peak location is in good agreement with previous observations of the shallow (~ 110 meV) N acceptor level.^{6,29} The important point in this series of spectra is that the $2S_{3/2}$ peak is strongest when it lies just on the high-energy side of the nonresonant DAP band, which is typical behavior for all SPL peaks.^{27,28}

The strength of the resonantly excited peak increases for more distant pairs (lower-energy PL energies) because they are statistically more common, assuming a random distribution of pairs in the lattice. However, more distant pairs also have longer radiative lifetimes, due to the reduction in the overlap of their wave functions; it is therefore more probable that the excitation will tunnel to other pairs prior to recombination, giving rise to non-selective rather than sharp selective luminescence. The optimal strength of the selectively excited peaks therefore occurs for large but finite pair separation, which typically corresponds to energies on the high-energy side of the nonresonant DAP peak.^{27,28} When the SPL peak lies on the lower-energy side of the nonresonant peak, it corresponds to very distant pairs and therefore its intensity drops dramatically, as evident in Fig. 5.

Figure 6 shows a similar series of SPL spectra for the more heavily doped sample No. 4, which shows both a “normal” shallow nitrogen DAP peak and the deeper ~ 2.678 -eV DAP peak for above-gap, nonresonant excitation. The same basic features are observed as in the spectra of the lower doped sample, but with one crucial difference. The $2S_{3/2}$ peak now remains strong at energies well below the peak of the shallow ~ 2.694 -eV DAP band as seen in the three lowest spectra, where the energy of the $2S_{3/2}$ is 2.6904, 2.6856, and 2.6802 eV, respective-

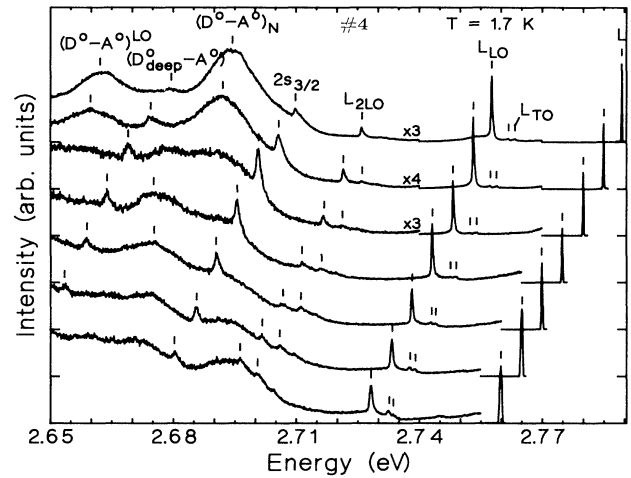


FIG. 6. As in Fig. 5, but for a more heavily N-doped sample exhibiting the deep DAP peak.

ly. The energy of the peak remains 79.5–79.7 meV below the laser line, however, indicating that it is still the ~ 110 meV acceptor level that is being excited. These observations show clearly that the peak associated with the usual shallow N acceptor level resonates across the position of the deep DAP peak. The SPL peak is absent for laser energies (not shown) of 2.755 eV and below, as would be expected based on the position of the nonselectively excited deep DAP peak. Very similar results have also been obtained for N-doped samples grown in other laboratories.²²

We also searched for evidence of the excited states of the donors involved in both the shallow and deep DAP peaks, but could not detect them due to the relatively high doping levels in these samples. Since the donor wave functions are much more extended than those of the deeper acceptors, their energy levels are more sensitive to broadening due to the random electric fields of the dopant ions. We detected the $2S$ states of ordinary shallow donors in more lightly N-doped material grown elsewhere, which exhibits only a single shallow DAP peak.²² In the present case, the observation of two distant DAP peaks, both involving the *same* 110-meV acceptor level, clearly implies that a deeper donor must indeed be present to produce the ~ 2.68 -eV pair peak. The SPL data thus provide positive confirmation of the original model presented for these peaks by Hauksson *et al.*⁸

D. Deep emissions

Figure 7 shows PL spectra for DAP emission recorded under different excitation intensities. We show three sets of measurements for the irradiated regions of three different samples. Figure 7(a) shows PL from sample No. 4, which has a net acceptor concentration of $2.1 \times 10^{17} \text{ cm}^{-3}$; Fig. 7(b) shows PL from sample No. 3 with net acceptor concentration of $2 \times 10^{17} \text{ cm}^{-3}$; and Fig. 7(c) shows PL from sample No. 2 with acceptor concentration of $4 \times 10^{17} \text{ cm}^{-3}$. The nitrogen flux during growth was lowest for sample No. 4 and highest for sample No. 2 and

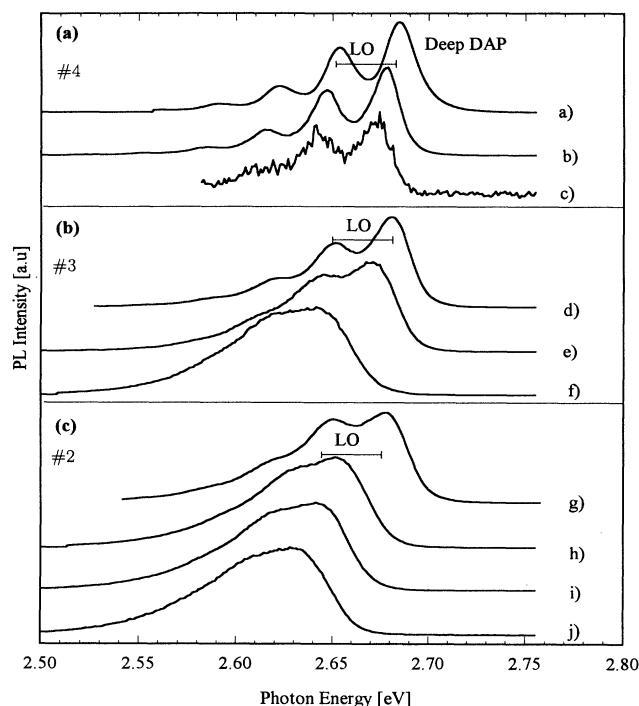


FIG. 7. PL spectra taken at 4 K showing deep DAP transitions for different excitation intensities taken inside the laser-irradiated region. Spectra (a)–(c) are for sample No. 4, spectra (d)–(f) for sample No. 3 and (g)–(j) are for sample No. 2. The excitation intensities are approximately (a): (a) 3.5 W cm^{-2} , (b) 0.1 W cm^{-2} , (c) $2 \times 10^{-4} \text{ W cm}^{-2}$; (b): (d) 3.5 W cm^{-2} , (e) 0.35 W cm^{-2} , (f) $3.5 \times 10^{-3} \text{ W cm}^{-2}$; and (c): (g) 1.0 W cm^{-2} , (h) 0.3 W cm^{-2} , (i) 0.05 W cm^{-2} , and (j) 0.01 W cm^{-2} .

so we expect the lowest nitrogen incorporation in sample No. 4 and highest incorporation for No. 2. In Fig. 7(a), the PL is dominated by deep DAP transitions and we observe a shift of the no-phonon line to higher energy with higher excitation intensity. This is explained in terms of recombination between close DAP's compared with distant pairs. The DAP recombination rate decreases exponentially with increasing separation, R . At low excitation, the emission occurs predominantly from relatively distant pairs, although competing recombination paths shift the peak to closer pairs than would be expected statistically.³⁰ When the excitation of the samples is increased, the recombination with large R saturates and the recombination with small R increases, that is, the DAP emission shifts to higher energy. Going from $1 \times 10^{-4} \text{ W cm}^{-2}$ to 1 W cm^{-2} the peak shifts up in energy by 11.8 meV while the shallow DAP shifts by 4.6 meV for the same intensity range. For low excitation intensity the Coulomb term is at a minimum since we are observing distant pairs and so we obtain the best estimation for E_D . Assuming that the Coulomb term is the same for shallow and deep DAP recombination, we get a new estimate of 46 meV as a lower limit for the activation energy of the deep donor.

For sample No. 3 [Fig. 7(b)], curve (d) shows the PL

spectrum for high excitation intensity and we can see the transitions corresponding to a deep DAP emission with phonon replicas. From curve (e) to (f) the excitation intensity is decreased and the emission shifts to lower energy and changes its shape. For sample No. 2 [Fig. 7(c)], the highest excitation spectrum (g) shows emission which is also close to the deep DAP transitions, but the peak on the lower-energy side does not correspond to the LO phonon replica. As we lower the excitation intensity the spectrum is characterized by two peaks, which are separated by approximately 23 meV. It is important to note that the shape of this deep emission does not seem to change when the excitation power is changed.

IV. DISCUSSION AND CONCLUSIONS

In Fig. 7, we compare three samples with almost the same active nitrogen concentration ($N_A - N_D$) but we observe different PL spectra due to the different degree of incorporation of nonactive nitrogen. Comparison by Qiu *et al.* of the ($N_A - N_D$), measured by $C-V$, with the nitrogen concentration $[N]$, as determined by secondary-ion-mass-spectroscopy (SIMS) shows that, typically, ($N_A - N_D$) reaches the value $5 \times 10^{17} \text{ cm}^{-3}$ quite quickly but then saturates as compensation by donors occurs. A typical value for $[N]$ for this saturation value is $1.8 \times 10^{18} \text{ cm}^{-3}$. The additional nonactive nitrogen may form donor complexes, which play a major role in changing the PL spectra. This kind of emission was first observed by Qiu *et al.*⁶ and they proposed that this emission is related to DAP recombination where the donor level has broadened into an impurity band. However, the excitation intensity-dependent PL measurements are not consistent with this model since the spectra move to higher energy with increased laser power consistent with DAP transitions.

The PL data at different temperatures show that two different donors are present in ZnSe with doping concentration around $1 \times 10^{17} \text{ cm}^{-3}$, a shallow donor with activation energy of 26 meV and a deep donor with activation energy of 46 meV previously proposed as a $V_{\text{Se}}\text{-Zn-N}_{\text{Se}}$ complex. In highly doped ZnSe ($> 2 \times 10^{17} \text{ cm}^{-3}$) the excitation laser power dependence of the PL shows an emission which is characterized by two bands separated by 23 meV without LO phonon replicas. The origin of these transitions is under investigation.

Finally, the selective pair luminescence measurements show that the same acceptor is involved in both the shallow donor-acceptor and deep donor-acceptor transitions, which is consistent with the temperature-dependent measurements on these emissions and is in agreement with the model of Ref. 8.

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