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Journal of Crystal Growth 159 (1996) 302–306

JOURNAL OF CRYSTAL GROWTH

Strain-dependent Zeeman effect of the nitrogen acceptor bound exciton in ZnSe-epilayers

A. Hoffmann [a,*], D. Wiesmann [a], I. Loa [a], R. Heitz [a], U.W. Pohl [a], I. Broser [a], L. Worschech [b], E. Kurtz [b], D. Hommel [b], G. Landwehr [b], D. Hoffmann [c], B.K. Meyer [c]

[a] *Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstrasse 36, D-10623 Berlin, Germany*
[b] *Physikalisches Institut der Universität Würzburg, Am Hubland, Würzburg, Germany*
[c] *Physik-Department E 16, Technische Universität München, Garching, Germany*

Abstract

We report on high resolution magneto-optical studies of the exciton bound to the neutral nitrogen acceptor in freestanding strain-free and strained ZnSe epilayers. The Zeeman results for absorption and photoluminescence are discussed in terms of the *j–j* coupling scheme in cubic symmetry including Zeeman and strain terms. It is shown that the bound-exciton state from the two hole $J=0$ state is below the bound-exciton states from the $J=2$ two-hole state. A diamagnetic shift of 5.6 $\mu eV/T^2$ is found. The g-value of the acceptor ground state is 0.7. The g-value of the bound-exciton state from the $J=0$ two-hole state is 1.35 representing the electron g-value. The g-value of the bound exciton from the $J=2$ two-hole state is 0.9.

1. Introduction

Nitrogen provides an efficient acceptor doping of ZnSe. The net acceptor concentration, however, is limited to about 10^{18} cm^{-3} due to compensation processes which are not yet well understood. Detailed information on the possible compensating defect is obtained from ODMR measurements of the donor acceptor pair (DAP) band in highly doped ZnSe:N [1,2]. The assignment of the observed resonances, however, is still controversial. Therefore, the purpose of this paper is to present detailed information on the electronic properties of both the N-acceptor and the (A^0_N,X)-complex. The well-defined nitrogen-acceptor–bound-exciton transition I_1^N in high quality MBE-grown ZnSe:N/GaAs-epilayers is studied by magneto-optical investigations. The ZnSe-epilayers are lifted from the GaAs substrate to get narrow I_1^N transitions (FWHM smaller than 200 μeV) and to allow absorption measurements. We compare Zeeman results for strained epilayers and freestanding strain-free epilayers.

2. Samples and experimental setup

The investigated ZnSe samples are high-quality p-type MBE-grown epilayers. The p-doping is performed with a nitrogen-plasma source. The two samples presented here are 0.96 and 3.8 μm thick with a net acceptor concentration of 3×10^{16} and 5×10^{15} cm^{-3}, respectively. For transmission experiments the ZnSe samples are glued onto quartzglass before the

* Corresponding author. Fax: +49 30 3142 2064; E-mail: axel0431@mailszrz.zrz.tu-berlin.de.

0022-0248/96/$15.00
SSDI 0022-0248(95)00816-0

GaAs substrate is removed by selective wet chemical etching [3]. The 3.8 μm thick sample has subsequently been released from the quartzglass for strain-free investigations.

Photoluminescence (PL) is excited by the 325 nm line of a He–Cd laser, dispersed by a 0.75 m double-grated monochromator and detected by a bialkali photomultiplier tube. For the monochromatic and polychromatic transmission spectra a tungsten lamp was used as excitation source. The Zeeman experiments are carried out using a superconducting 15 T magnet built in split-coil configuration at sample temperatures between 2 and 10 K.

3. Experimental results and discussion

The absorption spectra for strained and unstrained epilayers at 1.8 K are shown in Fig. 1c. In the strained epilayer a strain $\varepsilon = -2 \times 10^{-4}$ is determined from the free exciton splitting as described in Ref. [10]. Two zero phonon lines I_1^N and $I_1^{N'}$ are clearly resolved. This doublet can also be observed in emission with $I_1^{N'}$ occurring only at higher temperatures. Polarized excitation spectra of the (A_N^0,X)-complex allow to identify the excited fine structure states. The (A_N^0,X)-complex splits due to hole-hole-exchange interaction into the two-hole-state $J=0$ (origin of I_1^N line) and the $J=2$ level at higher energies (origin of $I_1^{N'}$ line). The additional coupling of the electron to the two holes within the (A_N^0,X)-complex results in a $j=\frac{1}{2}$ state originating from $J=0$ and states with angular momentum of $j=\frac{3}{2}$ and $j=\frac{5}{2}$ from $J=2$. These initial states can be described by hole–hole interaction $H_{hh} = a j_1 \cdot j_2$ and the electron–hole interaction $H_{eh} = b\boldsymbol{J} \cdot \boldsymbol{s}$ in a cubic crystal field V_c, j_i being the angular momenta of the two holes $j_1 = j_2 = \frac{3}{2}$ coupled to a total hole momentum $J=0$ and $J=2$ and the electron spin $s=\frac{1}{2}$.

The energy separation of I_1^N and $I_1^{N'}$ depends strongly on biaxial strain (Fig. 1a) and amounts to 0.28 ± 0.03 meV in strain free epilayers. This value is in excellent agreement with the corresponding fine structure splitting of the exciton bound to the neutral Li acceptor in bulk ZnSe [4]. Zeeman experiments up to 15 T were performed with both strain-free (Fig. 1b) and strained epilayers. For strain-free epilayers the Zeeman splitting of the absorption lines I_1^N and $I_1^{N'}$ in Faraday configuration for $\boldsymbol{B}\|[001]$ is presented in Fig. 1b. We observe four Zeeman components (two are polarized σ^+ and two σ^-). In Voigt configuration and $\boldsymbol{B}\|[110]$ two additional π-components are observed (in addition to the σ-components). In freestanding strain-free epilayers we found the Zeeman splitting to be isotrop within the experimental error. Neglecting anisotropic effects we determined from Fig. 1b the g-value of the acceptor $g_A = 0.70 \pm 0.09$, the g-value of the two-hole state $J=0$ $g_0^{BE} = g_e = 1.4 \pm 0.1$ and the g-value of the two-hole state $J=2$ $g_2^{BE} = 1.1 \pm 0.1$. In our transmission we are not able to resolve the fine structure of the $J=2$ two-hole state. The diamagnetic shift can be determined with 5.6 ± 0.1 μeV/T^2.

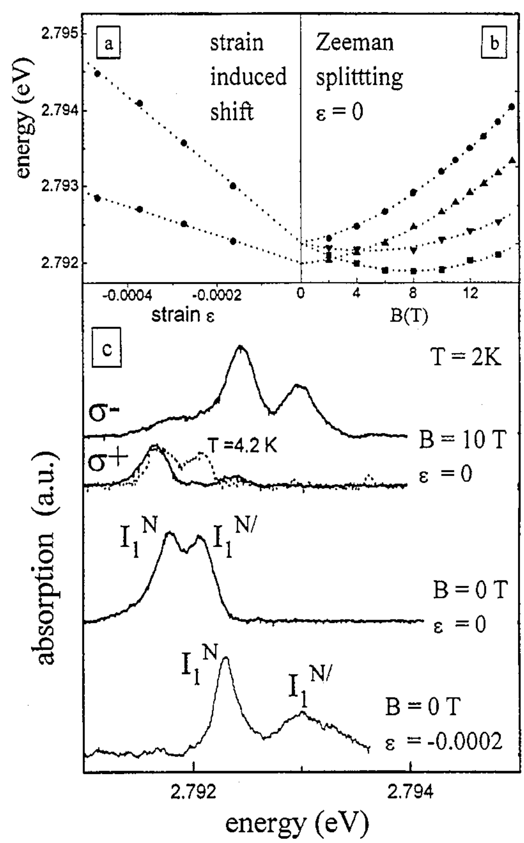

Fig. 1. (a) Dependence of the absorption lines I_1^N and $I_1^{N'}$ on the strain in ZnSe: N. (b) Zeeman splitting of the I_1^N and $I_1^{N'}$ bound-exciton line in Faraday configuration $\boldsymbol{B}\|[001]$. (c) absorption spectra showing the I_1^N and $I_1^{N'}$ lines at $B = 0$ and $B = 14$ T. The lower absorption spectrum shows the I_1^N and $I_1^{N'}$ lines in a strained ZnSe epilayer.

Fig. 2 shows the Zeeman splitting of the bound-exciton lines I_1^N and $I_1^{N'}$ in a strained ZnSe epilayer glued onto quartzglass. In addition to the Zeeman splitting for the magnetic field configurations $\boldsymbol{B}\|[110]$ and $\boldsymbol{B}\|[001]$ the angular dependence of the Zeeman splitting at 14 T is shown. Here, the Zeeman behavior exhibits a strong anisotropy. To analyse these complex data we have carried out a fit procedure taking simultaneously into account Zeeman and strain terms. For this purpose we combined the Hamiltonian of the magnetic field (as proposed by Molva et al. [5]) with an additional strain field Hamiltonian (as proposed by Schmidt et al. [6] and Laude et al. [7]). To fit the Zeeman splitting we assume the following model:

$$H_B = \mu_B\left[g_0 s\boldsymbol{B} + g_2 \boldsymbol{J}\cdot\boldsymbol{B} + Kj_1\cdot\boldsymbol{B} + L\left(B_x J_x^3 + B_y J_y^3 + B_z J_z^3\right)\right] + C_2 B^2,$$

with the Bohr magneton μ_B, Landé factors g_0 and g_2 of the electron and the coupled holes, respectively, the hole parameter K referring to the isotropic Zeeman effect, the anisotropic hole parameter L, and the diamagnetic shift parameter C_2.

For the strained ZnSe films we considered an additional planar strain ε which splits the $j_1 = \frac{3}{2}$ state into two branches commonly known as heavy and light hole. In the frame of the spin-Hamiltonian H_B the biaxial strain in the (001)-plane has been described as a small perturbation: $H_{strain} = \varepsilon_{strain}\,(j_z^2 - \frac{1}{3}j_2)$ acting on each hole j_i. Therefore, the magneto-optical data reveal the same g-factors for the strained and unstrained samples. The different Zeeman behaviour can be directly attributed to the strain-dependent term (ε_{strain}). However, while the effective g-factors $E = g_{eff}\,\mu_B mB$ differ for strained and unstrained samples, the electron g factor and hole parameters g_2 and K have been found to be unchanged whenever the strain is a small perturbation described by an additional term H_{strain} in the spin-Hamiltonian. Due to the coupling of the two-hole states to one state with angular momentum $J = 2$ all strain-dependent matrix elements vanish except the two:

$$\langle 0,0|H_{strain}|2,0\rangle = \langle 2,0|H_{strain}|0,0\rangle = \varepsilon_{strain}.$$

To describe the effect of strain on the Zeeman splitting we must additionally introduce a strain dependent electron–hole interaction $H_{\sigma e-h} = \sigma \boldsymbol{J}\cdot\boldsymbol{s}$, with the strain dependent electron–hole parameter σ.

The proposed Hamilton operator allows a good description of the experimental findings. Fig. 3 gives a fit of the angular dependence of the Zeeman splitting at 14 T. For the fit procedure two further lines $I_1^{N''}$ and $I_1^{N'''}$ have been taken into account.

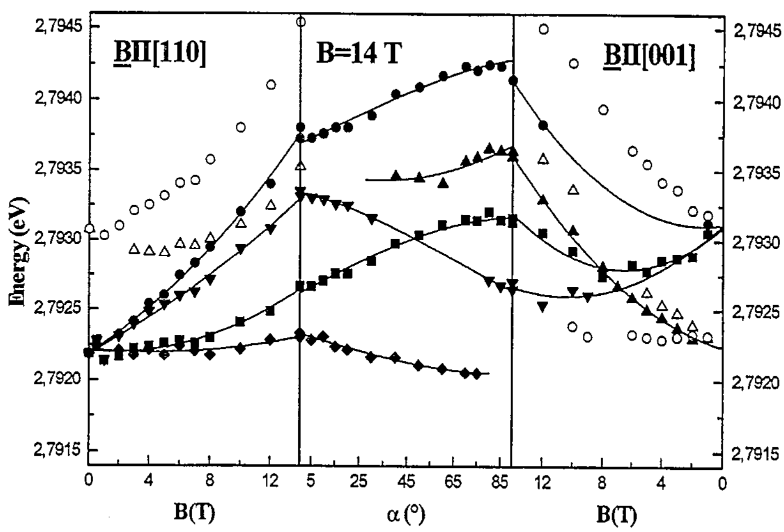

Fig. 2. Zeeman splitting of the I_1^N and $I_1^{N'}$ bound-exciton line in the configurations $B\|[001]$ (right) and $B\|[110]$ (left) in a strained ZnSe epilayer. The figure in the middle shows the angular dependence of the Zeeman splittings of the I_1^N and $I_1^{N'}$ lines at 14 T.

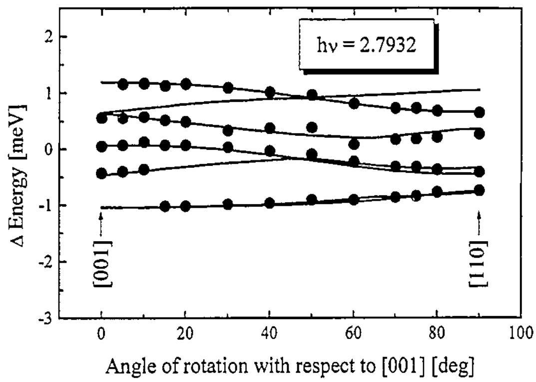

Fig. 3. The fitted (solid line) and observed (dots) angular dependence of the Zeeman splittings of the I_1^N and $I_1^{N'}$ lines at 14 T is represented.

These have been identified from quantum beat spectra observed in four-wave-mixing experiments for the I_1^N [8] as further fine structure splittings of the (A_N^0,X)-complex. In the investigated strained ZnSe epilayer we determined the energy separation between the I_1^N and $I_1^{N'}$ with 0.42 meV, the energy separation between the I_1^N and $I_1^{N''}$ with 0.65 meV and the energy separation between the I_1^N and $I_1^{N'''}$ with 1.27 meV. From the fit we received the following parameters: the isotropic hole parameter $K = 0.7 \pm 0.05$, the anisotropic hole parameter L is 0.03, the g-value of the $J = 0$ bound-exciton state $g_0 = 1.35 \pm 0.05$, the g-value of the bound-exciton state from the $J = 2$ two-hole state $g_2 = 0.9 \pm 0.05$, the parameter of the hole–hole interaction $a = 0.22 \pm 0.1$ meV, the parameter of the electron–hole interaction $b = -0.20 \pm 0.1$ meV, the cubic crystal field parameter $c = -0.14 \pm 0.1$ meV, the diamagnetic shift parameter $C_2 = 5.6$ μeV/T^2, the strain dependent electron–hole parameter $\sigma = 0.20 \pm 0.04$ meV, the strain parameter $\varepsilon_{strain} = 0.20 \pm 0.05$ meV.

For the two-hole state with $J = 0$ the g-value corresponds to that of the g-value of the bound electron, the value 1.35 is in good agreement with the g-value determined by Venghaus et al. [9] from magneto-optical investigations of the free exciton line in ZnSe bulk crystals. From ODMR measurements the g-value for the shallow donor in strained ZnSe epilayers is 1.15 [1,2]. This is seemingly a discrepancy to our results yielding 1.35 since in other II–VI compounds, like CdS, the electron g-values of the shallow donors and of the free and bound excitons are in the same order of magnitude. Our results show that for the bound-exciton states ($J = 0,2$) the Zeeman behaviour has a non-parabolic dependence and that the bound-electron g-value is influenced through the strong electron–hole interaction.

4. Conclusions

Our magneto-optical investigations give information about the fine structure of the bound-exciton states and of the nitrogen acceptor ground state in strained and unstrained ZnSe epilayers. The fit procedure of our Zeeman data is done in the framework of a *j–j* coupling scheme including Zeeman and strain terms. The g-value of the nitrogen acceptor is 0.7 independent of strain. The g-value of the bound-exciton state from the two-hole state $J = 0$ is 1.35 representing the electron g-value. The g-value of the bound-exciton state from the two-hole state $J = 2$ two-hole state is 0.9. Additionally, we determined the parameter of the hole–hole interaction $a = 0.22 \pm 0.1$ meV, the parameter of the electron–hole interaction $b = -0.2 \pm 0.1$ meV, the cubic crystal field parameter $c = -0.14 \pm 0.1$ meV, and the strain parameter $\varepsilon_{strain} = 0.2 \pm 0.05$ meV. Our Zeeman fitting gives clear evidence that the nitrogen behaves like an effective mass shallow acceptor. This indicates that it is incorporated on selenium sites.

Acknowledgements

Parts of this work were supported by the Deutsche Forschungsgemeinschaft.

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