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1997 J. Phys.: Condens. Matter 9 10795

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Shallow acceptors in GaAs grown from the vapour phase

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Received 23 June 1997

Abstract. The photoluminescent spectra from high-quality GaAs crystals grown from the vapour phase reveal a dominant Zn acceptor and a C acceptor, as well as an acceptor that would appear to have a smaller binding energy than C. We show that this shallowest acceptor is not a discrete chemical acceptor, having a smaller binding energy than C, but is rather due to a complex centre.

1. Introduction

A great deal of effort has been devoted to analysing acceptors in GaAs [1–14]. Selective pair luminescence (SPL) has been used to determine the excited state energies of a number of different chemical acceptors. There are some samples in which SPL is not observed. In particular, the samples reported in this paper do not exhibit SPL. The samples are vapour phase grown and are of quite good crystalline quality. In the energy region in which SPL would normally be observed, the excited state spectra of residual acceptors occur. The photoluminescence (PL) spectra are that of acceptor bound excitons (A_0 , X). The excited state spectra result when the exciton bound to the acceptor collapses from the initial state leaving the acceptor in an excited state in the terminal state. This is referred to in the literature as ‘two-hole’ transitions [1]. The excited state spectra in this study reveal a dominant Zn acceptor (1.490 55 eV) as well as a C acceptor (1.493 10 eV). In addition, one observes what appears to be an acceptor with a smaller binding energy than C (1.496 27 eV). A similar acceptor was observed by Contour *et al* [15] where it was identified as a shallow acceptor. It is the purpose of this paper to show that the acceptor we observe is not a unique chemical acceptor, having a smaller binding energy than C, but is rather a complex. The energy of the transition can be explained by a complex consisting of two acceptors and a donor.

2. Experimental details

The GaAs samples used in this study were undoped and of high quality. They were epitaxial layers grown on chromium-doped semi-insulating GaAs substrates by the $H_2:As:Cl_3:Ga$ vapour deposition technique. Transport measurements were used to characterize the electrical parameters of the samples. The samples were fairly compensated and may be either n or p type with total carrier concentrations $\sim 5 \times 10^{14} \text{ cm}^{-3}$. The high degree of compensation was reflected in relatively low mobilities.

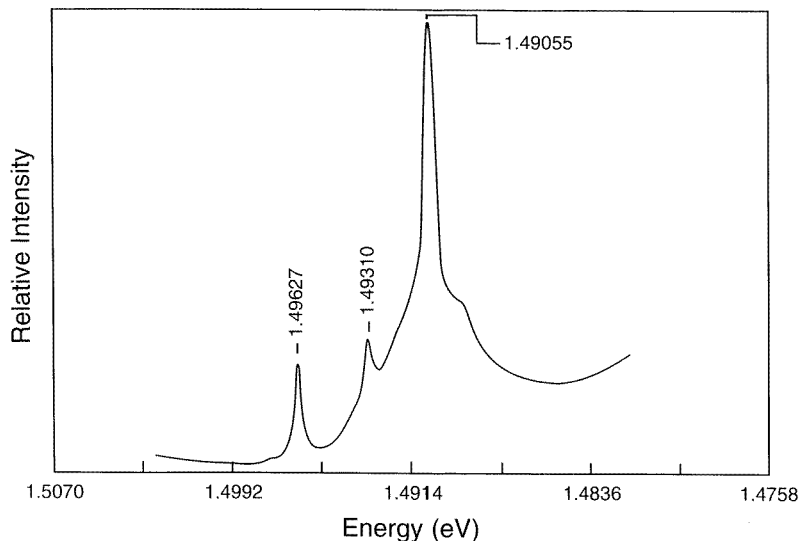


Figure 1. Energy positions of the Zn (1.490 55 eV), C (1.493 10 eV) and complex (1.496 27 eV) acceptors determined from PL.

The PL spectra were excited with an Ar⁺-ion laser-pumped tunable dye laser using Styryl 9 dye. The measurements were made at 2 K with the sample immersed in liquid He. The spectra were analysed with a high-resolution 4 m spectrometer equipped with an RCA C31034 Å photomultiplier tube for detection.

3. Experimental results

The spectra in the ‘two-hole’ excited energy state region, for one of the samples investigated, are shown in figure 1. The dominant transition associated with the Zn acceptor occurs at 1.490 55 eV, while the transition associated with the C acceptor occurs at 1.493 10 eV. The energies of these transitions are expressed as follows:

$$E_T = E_{ex} - E_A - E_A^* \quad (1)$$

where E_T is the transition energy, E_{ex} is the free exciton energy, E_A is the energy with which the exciton is bound to the acceptor and E_A^* is the energy required to put the acceptor in an excited state. The initial state of A^0, X consists of two $J = 3/2$ holes and one $J = 1/2$ electron. This spin configuration results in three initial states having J -values of $5/2$, $3/2$ and $1/2$. The energies of these states for Zn and for C are given in table 1. The details of the transitions from the initial to the excited terminal state for Zn and C are shown in table 2. The excited state energies are taken from Kisker *et al* [16]. It is noted that there is good agreement between the calculated and experimental energies.

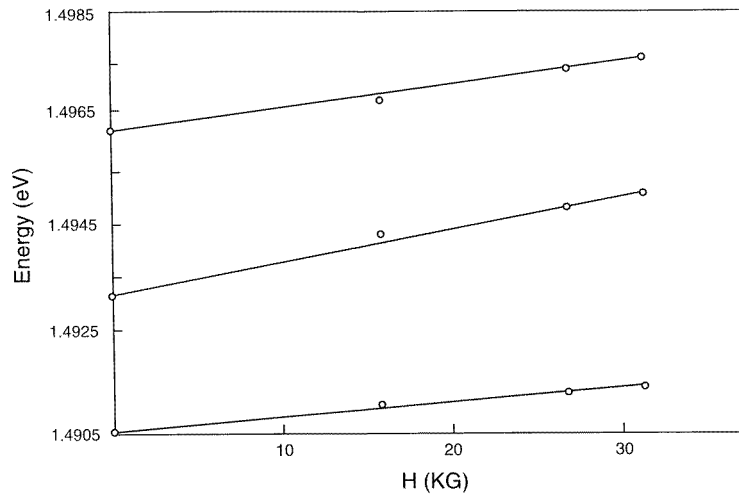
The transition at 1.496 27 eV in figure 1 might be interpreted as resulting from a discrete acceptor having a binding energy less than that of C (~ 23 meV). If this were the true source of this transition, then it should have a greater diamagnetic shift than the transition due to the C acceptor which has a greater diamagnetic shift than the transition due to the Zn acceptor. The diamagnetic shifts for these transitions are shown in figure 2. It is clear that the transition due to C has a greater shift than the transition due to Zn. It is also

Table 1. Energies of the excited states of Zn and C acceptors in GaAs.

Acceptor	Energy		
	$J = 1/2$	$J = 3/2$	$J = 5/2$
Zn	1.512 76	1.512 41	1.512 25
C	1.512 94	1.512 53	1.512 38

Table 2. Calculated and observed transition energies as well as the origin of the transition for GaAs.

Acceptor	Ground state	Excited state	Calculated energy (eV)	Observed energy (eV)
Zn	$1S_{3/2}$	$2S_{3/2}$	1.490 71	1.490 55
C	$1S_{3/2}$	$2P_{5/2} (\Gamma_8)$	1.493 13	1.493 10

**Figure 2.** Diamagnetic shifts of three acceptors shown in figure 1.

clear that the diamagnetic shift of the transition at 1.496 27 eV is comparable to or perhaps even slightly less than the diamagnetic shift associated with the C acceptor. This rules out the transition at 1.496 27 eV as being due to a discrete shallow acceptor having a binding energy of less than C.

Radiative transitions associated with complexes which involve two shallow acceptors and one shallow donor have been reported [17]. The model used to explain this complex is shown in figure 3. In the initial state there are two excitons, each bound to neutral acceptors, in the presence of a neutral donor. The exciton from acceptor 1 decays, giving up part of its energy to transfer the exciton from acceptor 2 to the neutral donor. It may also give up part of its energy to put the hole on acceptor 2 in an excited state. The terminal state will then appear as shown in figure 3. The transition energy will then be given by the following expression:

$$E_T = E_x - E_{A_1ex} - E_{A_2ex} - (E_{A_2} - E_{A_2}^*) + E_{Dex} \quad (2)$$

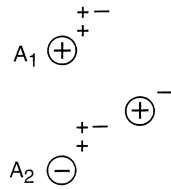
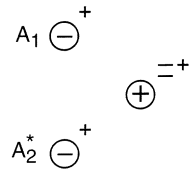
INITIAL STATE**FINAL STATE**

Figure 3. Initial and final states of the complex centre contributing to the shallowest acceptor shown in figure 1.

where E_T is the transition energy, E_x is the free exciton energy, E_{A_1ex} is the energy with which the exciton is bound to acceptor 1, A_{2ex} is the energy with which the exciton is bound to acceptor 2, $(E_{A_2} - E_{A_2}^*)$ is the energy required to put acceptor 2 into an excited state, and E_{Dex} is the energy with which the exciton is bound to the donor. The details of the transition from the initial to the final state resulting in the 1.496 27 eV transition in figure 1 are shown in table 3. In the first example the exciton collapses from the $J = 1/2$ state of acceptor 1, which is a C acceptor, removing the exciton from acceptor 2, which is also a C acceptor in a $J = 3/2$ state, and places it on the neutral donor. Acceptor 2 is also placed in a $2P_{3/2}$ excited state: again the excited state energy is taken from [16]. The calculated and measured energies of this transition agree very well. The good agreement suggests that the components of the complex are spaced relatively far apart and provides justification for neglecting the Coulomb terms in the calculation.

Table 3. Complex transition.

Collapsing exciton	Excited exciton	Terminal acceptor state	Donor state	Calculated energy (eV)	Observed energy (eV)
C- $J = 1/2$	C- $J = 3/2$	C- $2P_{3/2}$	D ₀	1.496 17	1.496 27
Zn- $J = 1/2$	C- $J = 1/2$	C- $2P_{3/2}$	D ₀	1.496 40	1.496 27

The other example shown in table 3 consists of a complex composed of one Zn acceptor, one C acceptor and a donor. In this example, the exciton collapses from the $J = 1/2$ state of the Zn acceptor 1, and removes the exciton from the C acceptor 2 which is in the $J = 1/2$ state. The exciton from C acceptor 2 is placed on the donor. In this case the calculated and observed transition energies agree almost as well as in the first example. There may well be other complexes that could also explain the transition observed at 1.496 27 eV.

In conclusion, the bound excitons in GaAs have a large spatial extent and therefore, even at low impurity concentrations, there is a good probability of observing transitions due

to complexes of the type described here. Since the energy of the final state is primarily that of the C acceptor in an excited state, it would be expected that the diamagnetic shift would be comparable to that of the 'two-hole' transition of the discrete C acceptor. This would be the case for the complex centres described here.

Acknowledgments

The authors express their sincere gratitude to C Huang and C W Litton for technical support. The work of DCR, DCL and BJ was performed at Wright Laboratory, Avionics Directorate (WL/AADP), Wright Patterson Air Force Base under USAF contract No F33615-95-C-1619. This work was partially supported by AFOSR.

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