Dynamical Behaviour of Excitons in ZnSe/GaAs Heterostructures

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The dynamical behaviour of free and bound excitons in ZnSe/GaAs epilayers grown by molecular beam epitaxy (MBE) was studied in the ps time range. From these investigations we determine the time characteristics of the excitation processes of the donor-bound excitons and the hh-lh conversion time within the free excitons to be 40-60 ps.

Introduction

Epitaxially grown ZnSe layers are the most promising II-VI compounds for optical and electrooptical devices, like the blue-light-emitting diode[1]. For the realization of ultrafast switching components, the precise knowledge of the dynamical behaviour of exited carriers is necessary. In this paper we investigated for the first time systematically the time dependence of the free- and bound-exciton luminescence in MBE-grown ZnSe/GaAs heterostructures.

Experimental

The samples were grown on (100) undoped GaAs substrates using a Perkin Elmar model 430 MBE system, see Ref. 2. For the time-resolved measurements we used an actively mode-locked Nd:YAG laser (Coherent) with a frequency-tripling BBO crystal pumping a dye laser.

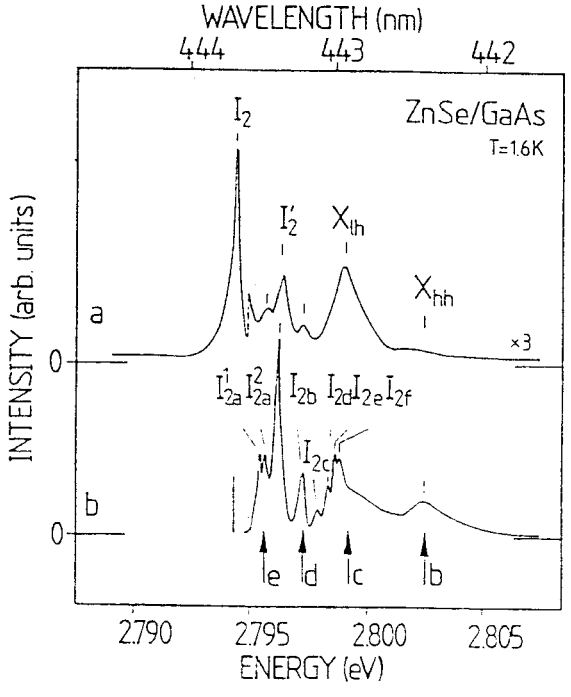

Fig.1
a) Luminescence spectrum of a 4 µm thick ZnSe/GaAs sample in the excitonic energy range, b) excitation spectrum of the donor-bound-exciton recombination line I_2. (arrows mark positions of detection of transients, see fig. 3)

The photoluminescence signal was analyzed by a single-photon-counting system and a micro-channel-plate photomultiplier tube.

Results and Discussion

In the time-integrated luminescence (upper part of Figure 1), the strain-split light- and heavy-hole free excitons X_{lh} and X_{hh} and two donor-bound exciton transitions $I_2^{(lh)}$ and $I_2'^{(hh)}$ are the dominant features. The I_1 line, related to the emission of the acceptor-bound exciton complex, is observed to be rather weak only.

Furthermore, a set of poorly resolved emission lines I_{2i} could be detected, which are much better resolved in the excitation spectrum of the I_2 (lower part of Figure 1).The I_{2i} lines were assigned to transitions from or into several excited single-hole states of the (D°,X) complex with different quantum numbers $|n,l>$ (see Ref.3).

In Fig.2, the transients of the free exciton lines X_{lh} and X_{hh} and of the donor-bound exciton lines I_2 and I_2' are shown for the case of nonresonant band-to-band excitation. The rise and lifetimes τ_r and τ_d of the excitons calculated from the fit of the transients are in the ps time range (see Table 1). For the analysis of τ_r and τ_d we used a sum of two exponential functions.

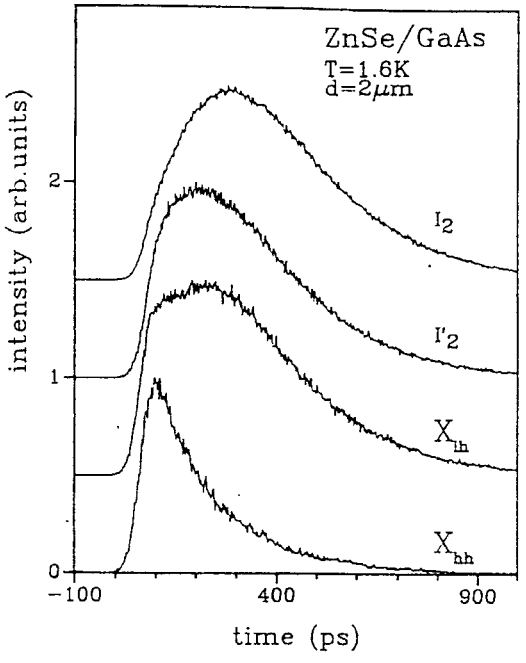

Fig.2
Time-dependent transients of the strain-split free excitons X_{lh} and X_{hh} and donor-bound-exciton transients $I_2^{(lh)}$ and $I_2'^{(hh)}$.

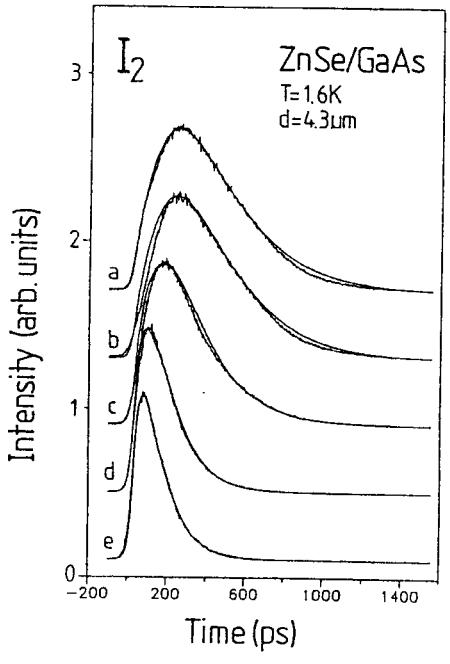

Fig.3
Transients of the I_2 recombination for different resonant excitation channels (see arrows in fig.1)

Table 1 *Measured rise times τ_r and lifetimes τ_d of different excitonic transitions in the case of nonresonant excitation.*

Line	X_{hh}	X_{lh}	I_2'	I_2	I_1
τ_r (ps)	20	80	110	150	120
τ_d (ps)	100	150	140	200	300

The observed rise times of the I_2' and I_2 emissions are correlated with the lifetimes of X_{hh} and X_{lh}, respectively. This confirms their assignment to the hh and lh components of the strain-separated ground state of the (D°,X) complex. The lifetime of the X_{lh} is significantly longer than this of the X_{hh} in accordance with their different oscillator strengths.
Additionally, the lifetimes of both free excitons are longer than that of the unsplit free exciton in unstrained bulk ZnSe crystals[4]. This may result from the high purity of the used epilayers.
The rise time of X_{hh} is considerably shorter than that of X_{lh}. After nonresonant carrier excitation, formation of free excitons obviously predominantly happens in the hh configuration after which a hh-lh coversion to the X_{hh} state may occur. The conversion time is 60 ps based on the given data.

To study the influence of strain in different layer regions[5] on the dynamics of the excitation we excited the I_2 line resonantly via the free excitons and several I_{2i} transitions. The used excitation energies are depicted in Figure 1 as arrows. In Fig. 3, the different I_2 transients are shown. In Table 2, the specific rise and lifetimes are given.

Table 2 *Rise times τ_r and decay times τ_d of the I_2 luminescence for different excitation situations*
E_G means nonresonant band-to-band excitation (see spectrum a in Fig. 1)

I_2	E_G	$E(X_{hh})$	$E(X_{lh})$	$E(I_{2b})$	$E(I_{2a})$
τ_r (ps)	150	130	90	40	30
τ_d (ps)	200*	200*	200*	100	80

* average lifetime of nonexponentially decreasing luminescence

For excitation beyond the band-gap energy E_G and with the energy of the free excitons X_{lh} and X_{hh} the lifetime of the I_2 emission remains constant, while the formation time τ_r changes drastically. This could be explained by energy relaxation from the band level to the free excitons and by an additional conversion time from X_{hh} to X_{lh}.
Concerning the I_2 decay times τ_d it is evident that, first, they considerably shorten when going from nonresonant or free-exciton resonant excitation to resonant excitation via excited (D°,X) states. Second, it is not possible to fit the transients exactly with only two exponential functions in the case of

nonresonant excitation. This may be explained by the fact that in this case (D°,X) complexes throughout the whole inhomogeneously strained layer are excited and all contribute to the observed decay characteristics. As strain affects the masses of the particles within the complex and, thus, the decay times, the I_2 luminescence is a superposition of components with different time constants[6].
In the resonant case, (D°,X) complexes in certain layer regions suffering from a specific strain magnitude are selectively excited. Consequently, the I_2 luminescence is no longer a superposition of differently decaying components, and reduced τ_r and τ_d are observed, corresponding to the real dynamic of the I_2 transition.

Table 3 *Rise times τ_r and lifetimes τ_d of the I_1 luminescence for different excitation situations*

I_1	E_G	$E(X_{hh})$	$E(X_{lh})$
τ_r (ps)	120	120	70
τ_d (ps)	300	300	280

Concerning the I_1 luminescence transients, we observed nearly unchanged lifetimes for different excitation conditions, whereas the formation time is strongly dependending on the excitation channel (see Table 3). Because τ_d is considerably shorter in the case of excitation via X_{lh} than via X_{hh}, the I_1 transition could be assigned to a lh correlated level of the (A°,X) ground state[7]. Generally, a difference of 40-60 ps is detected in the rise times of the I_1 and I_2 when changing the excitation energy from the X_{hh} exciton to the X_{lh} exciton transition. This value corresponds well to the difference between the exciton rise times (see Table 1). Therefore, we estimate the conversion between hh and lh correlated states to be in the range of 40-60 ps.

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References
1 M.A.Haase, J.Qiu, J.M.DePuydt, and H.Cheng, Appl.Phys.Lett. 59 (1991) 1272
2 R.L.Gunshor, and L.A.Kolodziejski, IEEE J.Quantum Electronics 24 (1988) 174
3 G.Kudlek, N.Presser, and J.Gutowski, Semiconductor Science&Technology.6 (1991) A83
4 G.Kudlek,U.W.Pohl, Ch.Fricke, R.Heitz, A.Hoffmann, J.Gutowski, and I.Broser, Physica (1992) in press
5 G.Kudlek, and J.Gutowski,Journal of Luminescence 52 (1992) 55
6 G. Kudlek, thesis, Technische Universität Berlin 1992
7 G.Kudlek, N.Presser, and J.Gutowski, Proc.eedings of the 21th ICPS, this issue