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Auger-type excitation and de-excitation processes in rare earth and transition metal doped semiconductors

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Abstract

Processes of indirect excitation of rare earth and transition metal ions are reviewed. We describe Auger-type processes, in which one of the interacting centres is ionised. We show that such Auger-type processes are often very efficient and result in indirect excitation of rare earth (transition metal) emission. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Semiconductors; Impurities in semiconductors; Recombination and trapping; Optical properties; Electron paramagnetic resonance; Luminescence

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1. Introduction

Several commercial light emitting devices were constructed based on wide band gap semiconducting materials doped with rare earth (RE) or transition metal (TM) [1,2]. In the case of RE-activated devices the crucial problem is the low efficiency of direct intra-4f-shell excitation of RE ions. Not surprisingly, the search for the appropriate indirect RE excitation mechanisms was very active in recent years. Several indirect RE excitation and de-excitation processes were shown to be important [1] and some of them will be analysed here. The first process discussed by us relates to RE excitation caused by energy transfer from adjacent donor–acceptor pair (DAP) to RE (TM) ion. In most of the cases the process leads in intra-shell excitation of RE (TM) ion. However, this process and also the reverse process of RE(TM)-to-DAP energy transfer can be of the Auger-type, as will be explained later.

The second indirect excitation mechanism discussed in the present work relates to the binding of excitons by RE (TM) ions. Several results suggest that exciton binding, followed by intra-shell excitation of RE (TM) ion, may be a quite common RE (TM) excitation mechanism [1]. Difficulty of its observation relates to a very high probability of nonradiative recombination of such excitons. For most of the cases exciton bound by RE (TM) ion decays nonradiatively and energy is transferred to core states of the impurity, which was binding the exciton. The energy transfer results thus in intra-shell excitation and not in ionisation of RE (TM). Despite this fact, such transfer process is often called the internal Auger process. Therefore, we will include to the present work discussion of this process, in which we describe Auger-type processes of indirect RE (TM) excitation. Only in few cases the energy transfer from bound exciton to impurity results in ionisation of the ion, which was binding the exciton.

Processes of RE (TM) ionisation will also be analysed. We will show that carrier trapping by ionised impurity can proceed by one of the excited core states and thus result in intra-shell emission of RE (TM) ion. RE (TM) ionisation, which e.g. can be induced by Auger-type energy transfer from DAP, can thus result in indirect excitation of RE (TM) intra-shell emission.

Some of TM impurities, when introduced to wide band gap II–VI materials, act as efficient centres of nonradiative recombination. Visible DAP emission is deactivated, which in most of the cases is due to DAP recombination transitions. In addition to the so-called bypassing process discussed in detail in the reference [3], recent photoluminescence (PL) and electron spin resonance (ESR) studies indicate the importance of the Auger-type nonradiative process, in which energy of recombining donor–acceptor pair is transferred to a nearby TM ion, causing its ionisation. This Auger-type nonradiative process was first observed for Fe ions in ZnS:Cu [3] and then for Fe and Cr ions in ZnSe [4]. The efficiency of this process will be discussed here based on the results of our recent ESR and PL investigations of undoped and chromium or iron doped ZnSe crystals. We will show that the Auger-type energy

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transfer from DAP-to-TM ion is the most efficient in samples doped with iron.

The role of Auger-type processes in low-dimensional structures is much less studied and understood. We will discuss here the first results of PL and time-resolved PL investigations of (CdTe,CdCrTe)/CdMgTe quantum well (QW) structure grown by molecular beam epitaxy. The observed strong reduction of PL intensity and decay time in chromium doped QWs is explained in terms of Auger-type energy transfer from excitons to chromium ions. We find that the Auger process is more efficient than the bypassing process, which is the opposite situation to that observed in bulk samples.

2. Indirect mechanisms of RE and TM excitation

2.1. DAP-to-RE energy transfer

Importance of this indirect excitation process of RE ions was realised quite early [5]. For wide band gap II-VI luminophors this process turned out to be the most important and thus was the most widely studied mechanism of indirect excitation of RE PL. The evidence for DAP-to-RE energy transfer was reviewed in details by other authors [5–9] and will not be given here. Energy transfer from an excited DAP to RE ions was observed in practically all wide band gap II–VI materials. The theoretical description of the process was developed by Schaffer and Williams [6], who related the transfer efficiency to the spectral overlap between DAP emission and RE absorption.

In their approach, Schaffer and Williams considered the multipole mechanism of the energy transfer [6]. In such a case, energy transfer should proceed to relatively distant RE centres. However, it was then demonstrated experimentally that DAP-to-RE transfer is only efficient for these RE ions which are closely associated with one of the components of DA pair. For distant pairs this process is apparently less efficient [9].

2.2. RE-to-DAP energy back-transfer

Spectral overlap between RE emission and DAP absorption means that energy back-transfer from excited RE ion to DAP can also be important. Considering rather low efficiency of 4f–4f intra-shell radiative recombination the process of the energy back-transfer can be quite destructive. Surprisingly, until recently such energy back-transfer was not observed experimentally.

The obvious consequence of the energy back-transfer is shortening of the decay time of RE PL. Thus, the efficiency of the energy back-transfer should be estimated from studies of PL kinetics. However, this estimation turned out to be experimentally difficult. It is usually quite difficult to conclude if the observed PL decay time is purely radiative or if it is shortened by the competition of nonradiative transitions, including the energy transfer processes mentioned above. The discussion of PL decay times complicates the fact that the probability of PL transitions strongly depends on the local symmetry of various RE complexes.

To verify the role of RE-to-DAP energy back-transfer we studied Yb^{3+} emission in InP and in alloys of InP with InAs, were we could change spectral overlap between RE PL and DAP absorption by changing the alloy composition. For the InAsP samples with 4 and 7% fractions of As, the Yb^{3+} PL is superimposed on a low energy wing of the DAP band. The overlap increases with increasing As fraction in the alloy. Thus, the energy back-transfer from excited 4f state of Yb^{3+} to DAP band is expected to be enhanced with increasing As fraction and the efficiency of RE-to-DAP nonradiative transition should be increased. Moreover, a fairly simple Yb^{3+} PL is observed in InP and in the alloys. Whereas for other RE ions several RE complexes are active in PL transitions (see e.g. [9]), only one Yb centre is observed in the PL of InP. In consequence, analysis of PL kinetics is more straightforward, since we do not compare PL decay for different Yb^{3+} sites.

In Fig. 1 we show the PL kinetics of the Yb^{3+} 4f–4f emission in InP and in two samples of InAsP. The decay time of the 4f–4f PL of Yb^{3+} clearly depends on As fraction in the alloy. It is (at 4.2 K) 10.4 μs in InP but rapidly shortens in the alloys. It is 4 μs for the 4% and 2.5 μs for the 7% As fraction in InAsP [10]. We believe that RE-to-DAP energy back-transfer is important and is responsible for the observed shortening of the PL decay time [10].

The relevant question is what is the mechanism of the RE-to-DAP energy back-transfer. For InP and in its alloys with InAs ionisation energy of shallow donors, which participate in DAP transitions, is very small. The observed spectral overlap between Yb^{3+} PL and DAP PL means thus that such overlap occurs also between Yb^{3+} PL and DAP absorption (DAP excitation). DAP PL is induced by ionisation transitions of acceptors. Acceptor centres are ionised and electrons are excited to conduction band. Photo-generated free electrons are then trapped by donor centres participating in the DAP recombination. In consequence, we propose that Yb-to-DAP energy back-transfer is of the Auger-type, i.e., Yb decays nonradiatively due to the Auger-type energy transfer to acceptor centres resulting in their ionisation.

2.3. Exciton binding by RE (TM) ions

Simple quantum mechanical considerations indicate that practically any foreign atom replacing host atom can introduce a short-range binding potential for either an electron or a hole. This is due to a difference in electronegativity of impurity and host atom or/and difference

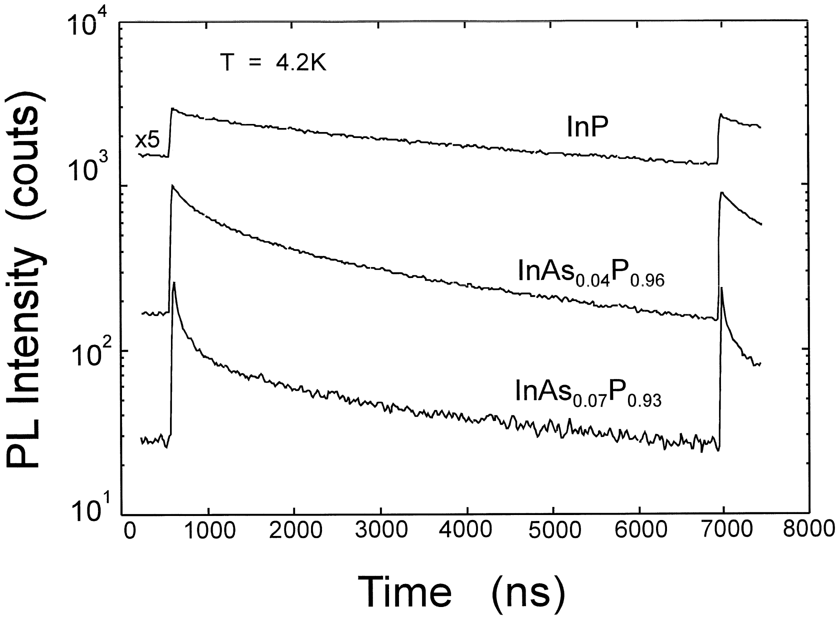

Fig. 1. Low temperature (4.2 K) PL decay times of the Yb^{3+} 4f–4f emission in InP and in two InAsP alloys with 4% and 7% As fraction.

in strain conditions between undoped and doped crystal. Presence of a short-range binding potential results in localisation of a first carrier (electron or hole, depending on the nature of the potential) by the impurity. Long range Coulomb potential centred at the impurity site is then created and binding of a second carrier (of an opposite sign) becomes very efficient. This scenario explains why excitons can be trapped. Thus, one may expect that exciton binding by RE and TM centres can be very probably. However, we will further show that for most of RE ions the trapping rate of free carriers is unfortunately low. This partly explains why exciton binding was discussed for several of TM ions, but is considered to be important only in very few cases for RE ions [1,11].

Even though the exciton binding was suggested for several of RE ions [1,10–14] the direct proof of it is still missing. This comes from a high probability of energy transfer from exciton to impurity core states, i.e., from a high probability of internal Auger process mentioned in the Introduction. Exciton binding and the following energy transfer from RE bound exciton to RE 4f core states (internal Auger effect) were discussed in detail in the references [1,11]. For RE (TM) ions energy for intra-shell 4f–4f (3d–3d) excitation is usually similar to that of recombining exciton. This makes internal Auger process highly efficient, but also means that a so-called satellite emission of bound excitons is not observed. By satellite PL of bound excitons we mean PL induced by a transfer of a part of exciton energy to core states of centre binding the exciton [15]. Satellite PL of RE (TM) bound excitons can thus be observed when there is a large energy mismatch between exciton emission energy and core excitation energy. Till now the ‘‘satellite’’ emission was only observed for Yb ion in ZnS [16].

The important property of satellite exciton lines is that the lines due to parity forbidden s–s transitions of neutral donor (acceptor) are strong [15]. This is due to lack of the rotational symmetry in a complex consisting of three carriers, electron and hole from an exciton and a carrier bound at neutral donor (acceptor). Similar effect is expected in the case of RE (TM) bound excitons, which may explain why the energy transfer from RE (TM) bound exciton to impurity core states is so efficient and results in an efficient 4f–4f (RE) or 3d–3d (TM) intra-shell excitation.

2.4. RE-to-exciton energy back-transfer

For all the cases with only small energy mismatch between exciton energy and energy required for RE (TM) core excitation, the process of exciton-to-RE energy transfer should be very efficient. It is also true for InP:Yb [12–14]. The small energy mismatch means, however, that the reverse process, i.e., energy transfer from excited RE ion to exciton can also be efficient at higher temperatures. In fact, intensity of the Yb^{3+} PL and PL decay time of 4f–4f emission decrease at increased temperature, which is explained by the energy back-transfer from the excited 4f state of Yb to either the Yb-bound exciton [13] or to band gap states of InP [17].

To verify the relative importance of these two possible back-transfer processes we studied the temperature dependence of the Yb^{3+} PL in InP and in InAsP [10]. Our experimental results confirm that the energy back-transfer is efficient at increased temperatures. PL intensity is reduced at increased temperatures. For InP we found that the PL deactivation energy is about 190 meV, which, after introducing a $T^{3/2}$ correction to the pre-exponential term, is equal, within the experimental error, to the energy difference between InP band gap energy and the Yb^{3+} PL emission energy. Deactivation energy of 145 meV was expected in that case [13]. Thus, the observed energy back-transfer is of the Auger-type, since nonradiative decay of Yb^{3+} PL results in band-to-band excitation. For the 7% sample the PL deactivation energy was 50 meV, once more confirming our conclusion on the energy back-transfer to band states [10]. Similar conclusions were also drawn from measurements of PL response to microwave power, which are described elsewhere [10,18].

2.5. Ionisation transitions of RE ions

Several of the experimental results indicate that some of the RE ions and the majority of TM ions can change their charge state in wide band gap materials [1,19,20]. In Fig. 2 we show theoretical estimation of ionisation energy required for 3+ to 2+ transitions of RE ions in wide band gap II-VI sulphides. The solid line in Fig. 2 represents results of our semi-empirical calculations [1,19,20], which were performed using refined spin-pairing model of Jörgensen [21]. Energies of 3+ to 2+ ionisation were calculated treating ionisation energy of free RE ion (reduced in the crystal by electrostatic interaction with ligands) and energy of interaction between 4f electrons as fitting parameters [19,20]. The theoretical treatment is described in the reference [19]. The predicted ionisation energies should now be compared with band gap energies of several wide band gap II–VI compounds. Such comparison indicates that in the case of CdS two of the RE ions (Eu and Yb) can change their charge state from 3+ to 2+, whereas in ZnS four of the RE ions (Sm, Eu, Tm and Yb) can change their charge state. For wider band gap CaS and SrS few further RE ions can be ionised. These theoretical predictions were confirmed experimentally for ZnS, CaS and SrS, as is indicated in Fig. 2. The most reliable proof of RE ionisation was obtained from ESR investigations [16,22].

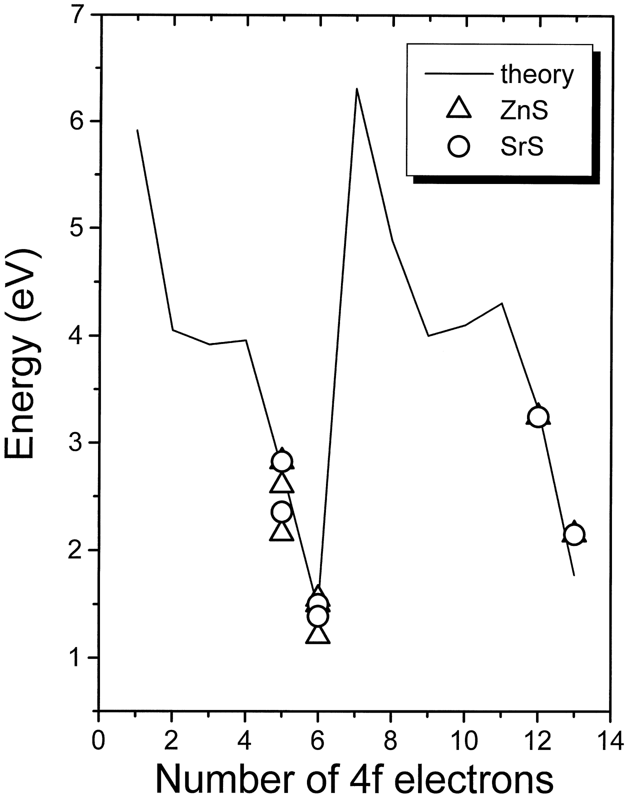

Fig. 2. Ionisation energies of RE ions in wide band gap sulphides. Solid line represents the results of semi-empirical calculations performed by us [19] using Jörgensen theory [21]. Open circles and triangles represent experimental results, which are summarised in the reference [1].

2.6. Localised bound exciton states of RE ions

Possibility of RE (TM) ionisation results in several new indirect RE (TM) excitation processes. All of them require that ionised carrier is either directly or indirectly trapped via excited 4f (3d for TM) or 5d state of RE. For CaS the electron is directly trapped to $4f^6 5d^1$ excited state of Eu^{2+} [23]. However, in the case of ZnS:Eu such a state is resonant with conduction band. In this case, the carrier is trapped via extended excited state of the Eu ion [1,23]. Such an extended state is called an impurity charge transfer state of RE (TM) ion or a localised bound exciton state [1]. Formation of RE (TM) localised exciton state was suggested for several of RE and TM ions [1,22–24].

2.7. Three-centre Auger excitation of RE emission

In this section we will describe another process, which relates to RE (TM) ionisation. We will analyse an Auger-type energy transfer process, in which RE (TM) ion is ionised by energy transfer from adjacent DAP. In this sense the process is an analogue of DAP-to-RE transfer process discussed above and is a close analogue of the three-centre Auger process discussed previously for some of the TM ions [3,4,24–26] and for shallow donor centres [27].

In the DAP-to-RE energy transfer process, as was discussed in the Section 2.1, DAP decays nonradiatively and the DAP recombination energy is ‘‘used’’ for intra-shell excitation of RE ion. In the ‘‘new’’ transfer process recombination energy of DAP is ‘‘used’’ for RE ionisation. Absorption bands due to ionisation transitions are much broader than those due to RE intra-shell transitions. This increases the chance of spectral overlap between DAP emission and RE absorption (ionisation). Moreover, absorption due to ionisation transitions is more intense than 4f–4f absorption. Considering these facts DAP-to-RE transfer is expected to be fairly efficient.

In Auger-type DAP-to-RE (TM) energy transfer three different centres participate. Therefore, the process can be called three centre Auger recombination (TCAR) [24,25]. The TCAR process was first demonstrated from ESR studies of Fe ions in ZnS [3] (see Fig. 3). Then, the process was proposed for other TM ions in ZnS and ZnSe [4,24–26]. This process was also proposed to explain indirect excitation of Yb^{3+} ion in ZnS lattice [16]. We expect that the TCAR process can be important for all RE and TM ions, which can change their charge states in a given lattice.

However, for a high overall efficiency of RE intra-shell excitation it is crucial that ionised RE ion efficiently traps carrier. Moreover, carrier trapping must proceed via one of the excited RE states. Unfortunately this is not the case. Our ESR studies of Eu ion in ZnS show rather low efficiencies of carrier trapping by ionised Eu ions [22]. We studied time evolution of ESR signals of Eu^{2+} and Cr^{1+} ESR signals under illumination, which generates free electrons and holes. Whereas Cr^{1+} ESR signal is rapidly quenched by illumination creating free holes (hole trapping results in 1+ to 2+ transition of chromium ions) Eu^{2+} ESR signal remains practically unchanged, proving a low efficiency of 2+ to 3+ transitions of europium ions [22]. Similar situation occurs also for Yb centres in ZnS, which suggests that low trapping efficiency is a common property of all ionised RE ions. Moreover, this efficiency can be further reduced in electroluminescence devices by the presence of strong electric field in active part of the device [1].

3. Three-centre Auger recombination in TM doped ZnSe

We performed the detailed studies of TCAR process for iron and chromium doped ZnSe. For both these TM ions we could observe photo-sensitive ESR signals. We could thus directly verify the role of TCAR processes in deactivation of visible emission of ZnSe, by studying spectral dependencies of photo-sensitivity of ESR signals (see Fig. 4). In this way we determined the nature of the observed absorption bands and we identified absorption bands due to ionisation transitions of Cr and Fe ions, and of deep ZnSe acceptors. We could thus directly prove that there is a spectral overlap between DAP emissions and Cr, Fe ionisation absorption.

We found that both ESR signals are photo-excited once DAP emission is excited, i.e., when deep acceptors are photo-ionised (see DAP band indicated in the Fig. 4). Once acceptors are ionised, free electrons are photo-created in

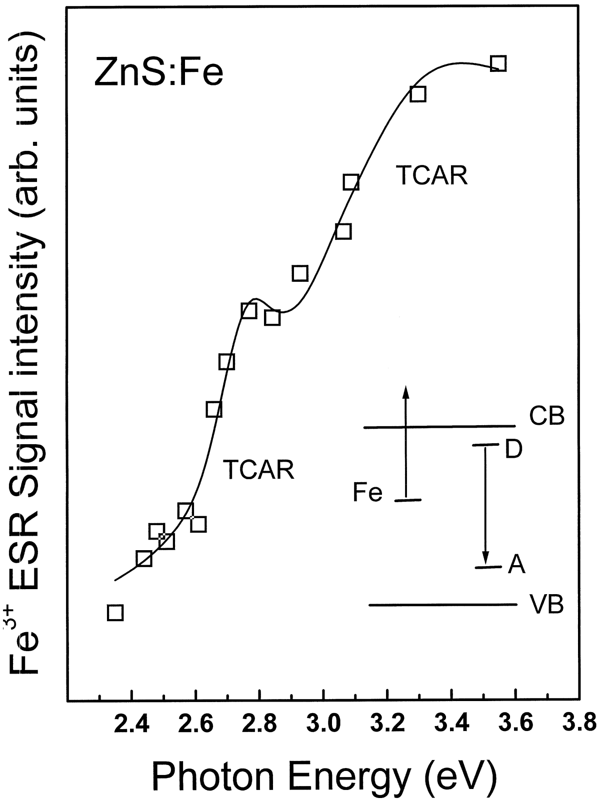

Fig. 3. Spectral dependence of photo-sensitivity of Fe^{3+} ESR signal in ZnS. By TCAR we indicate indirect excitation bands of Fe 2+ to 3+ ionisation, which are related to Auger-type energy transfer from DAP to iron centres, shown schematically in the figure.

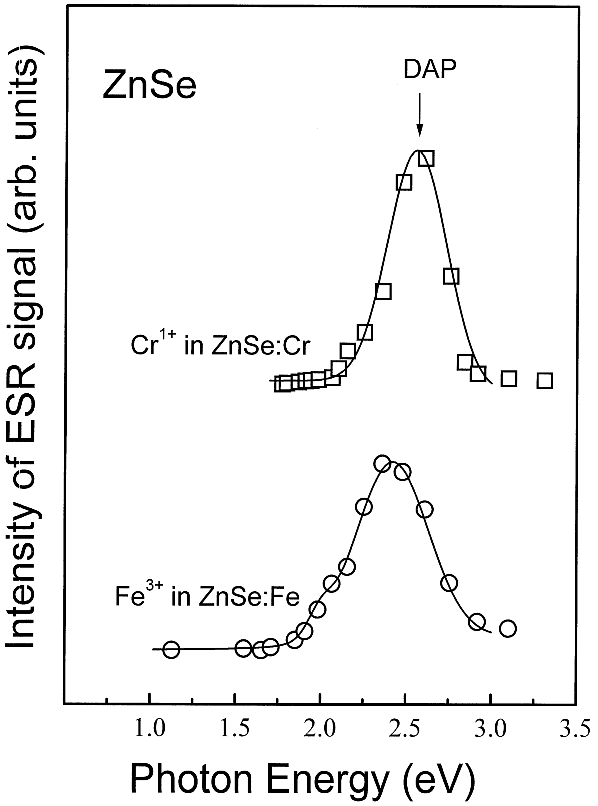

Fig. 4. Spectral dependencies of photo-sensitivity of Cr^{1+} (upper curve) and Fe^{3+} ESR signals in ZnSe. By "DAP" we denote excitation bands, which are due to ionisation transitions of deep ZnSe acceptors.

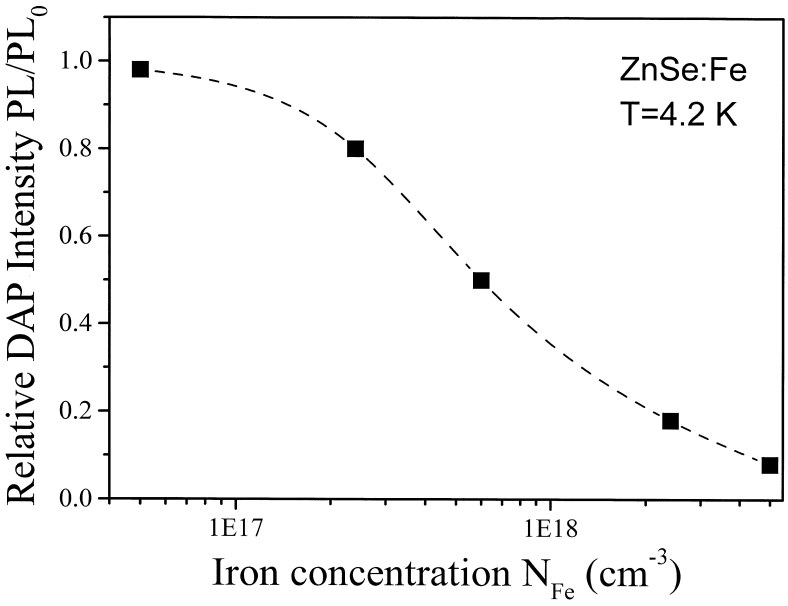

Fig. 5. Dependence of PL intensity of red DAP emission of ZnSe on iron concentration in the sample.

conduction band of ZnSe. These electrons can then be trapped by Cr^{2+} ions, which can explain photo-excitation of the Cr^{1+} ESR signal. Photo-sensitivity of Cr^{1+} ESR signal can thus be simply explained by carrier trapping processes (bypassing process) and the role of the alternative TCAR process remains unknown.

However, photo-generation of free electrons should result in photo-quenching and not in photo-excitation of the Fe^{3+} ESR signal. We explain photo-generation of this ESR signal by the TCAR process, in which free electrons are first trapped by shallow donors and then participate in DAP recombination, which can be nonradiative, due to the Auger-type energy transfer to iron centres. In this way, we could directly proof relatively high efficiency of the TCAR processes both in ZnS (see Fig. 3) and ZnSe (Fig. 4). Our ESR studies were supported by PL investigations. We found that both Fe and Cr act as efficient centres of nonradiative recombination in ZnSe, reducing intensity of visible DAP PL, as is shown in Fig. 5 for red DAP emission in iron doped ZnSe. This figure shows that intensity of red DAP PL is noticeably reduced in samples codoped with iron. The TCAR process must thus be relatively efficient [3,4].

We have also studied intensity, dynamics and spectral shape of free-to-bound (FB) and “shallow” DAP PL transitions (see Figs. 6–8). Due to relatively small donor

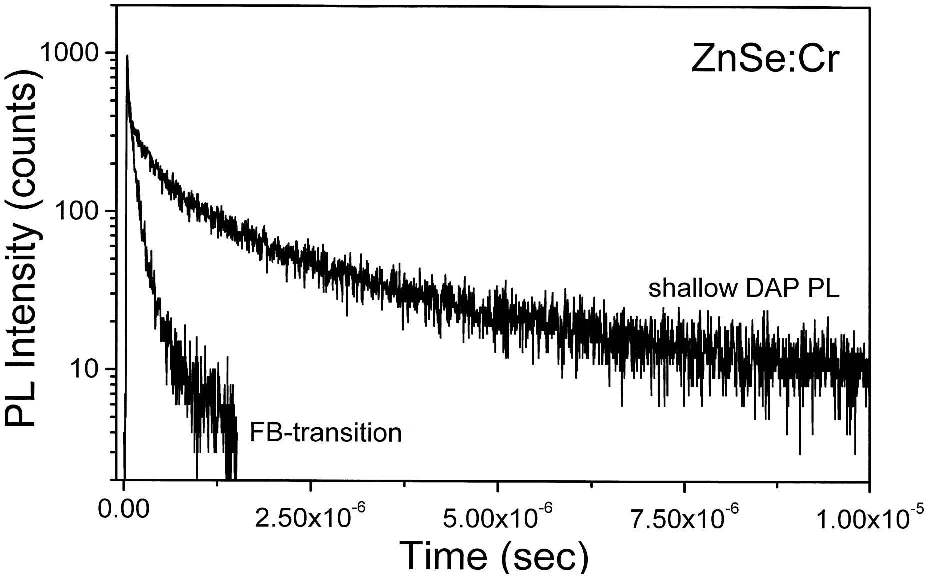

Fig. 6. PL decay times of FB (free electron-to-shallow acceptor) and “shallow” DAP emissions of ZnSe in chromium doped sample.

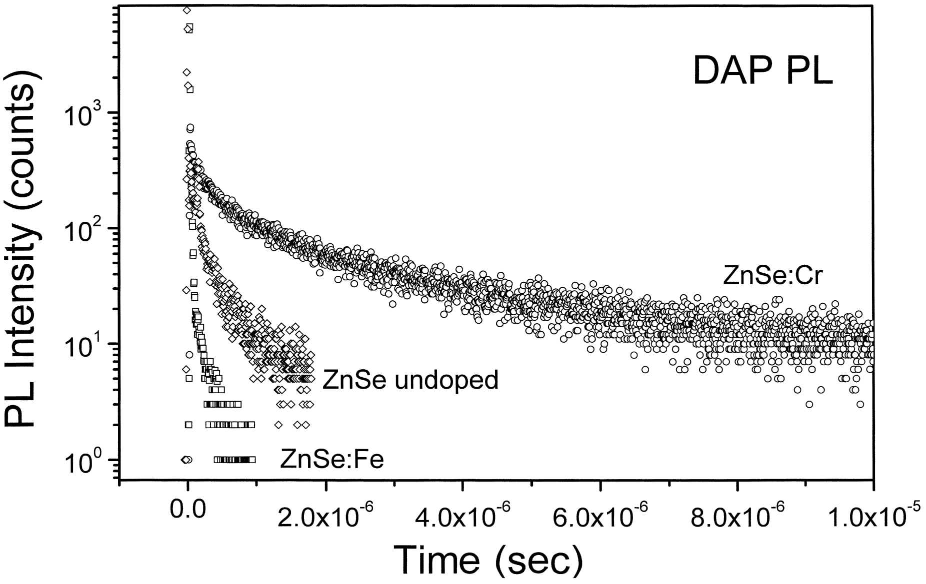

Fig. 7. PL decay times of overlapping FB (free electron-to-shallow acceptor) and ‘‘shallow’’ DAP emissions of ZnSe in undoped, chromium doped and iron doped ZnSe samples.

binding energy in ZnSe FB and ‘‘shallow’’ DAP emissions spectrally overlap and their relative intensities depend on temperature or, in time-resolved PL investigations, on time delay after excitation. The latter is possible since FB PL shows much shorter decay time (Fig. 6). The main effect of Cr codoping is the reduction of contribution of FB transitions to the ‘‘edge’’ PL. This is why the fast component of decay of the ‘‘edge’’ PL intensity is weaker in this sample, as can be seen in Fig. 7. Such reduction of the FB PL we explain by the so-called bypassing process, which was discussed in Refs. [3,4,25].

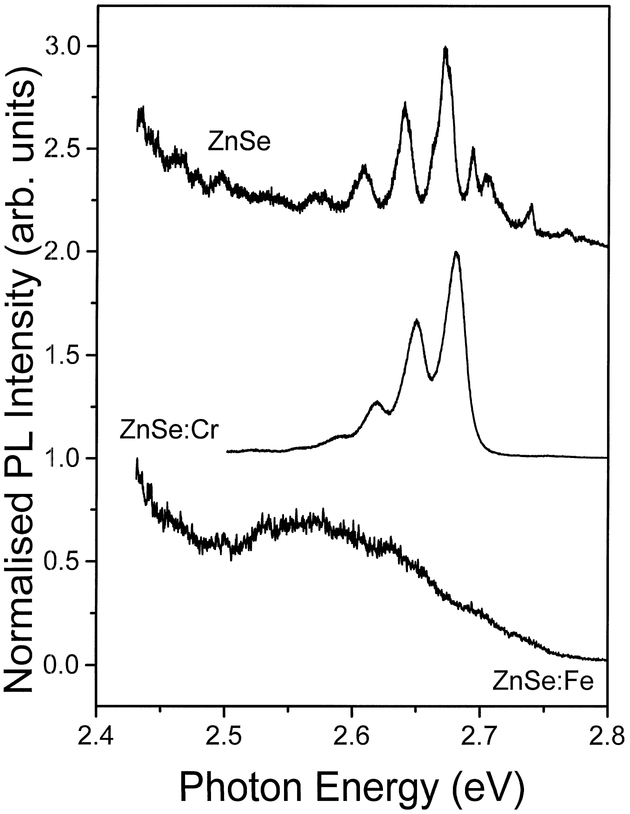

Fig. 8. ‘‘Edge’’ PL emission, due to overlapping FB (free electron-to-shallow acceptor) and ‘‘shallow’’ DAP transitions of ZnSe in undoped, chromium doped and in iron doped samples.

The very different situation we find for Fe doped sample. Here both ‘‘edge’’ PLs are very weak, broad and featureless (Fig. 8) and the PL decay time is noticeably reduced by nearly one order in magnitude (Fig. 7). These results indicate the high efficiency of both the bypassing process and the Auger-type energy transfer process. High efficiency of the latter process was already concluded from our photo-ESR studies of Fe^{3+} ESR signal [4].

The obvious question is why the Auger-type process is efficient in Fe but not (less?) in Cr doped sample. Formally, the theory of Schaffer and Williams [6] predicts that the transfer efficiency should only depend on spectral overlap between DAP emission and TM ionisation absorption. If so, the Auger processes in Cr and Fe doped samples should be of a similar efficiency, since ionisation transitions of Cr and Fe occur at similar energies. This definitely is not the case. Fig. 8 shows that the ‘‘edge’’ emission in iron doped ZnSe is not only very weak, but additionally is broad and featureless. Moreover, the ‘‘edge’’ PL lines do not narrow at longer delay times in time-resolved PL experiment, as was observed in Cr doped sample. We explain such features of the ‘‘edge’’ PL by effects related to a close association of iron with one of the component of DA pairs. High probability of formation of iron-acceptor associates we already concluded from previous optically detected magnetic resonance studies [28]. We thus propose that the three centre Auger process is so efficient since Fe ions are closely associated with DAP components. This is similar observation to the one given above on the efficiency of DAP-to-RE (intra-shell) excitation.

4. Auger mechanism of TM ionisation in quantum well structures

Recently we have studied recombination processes in 2D structures containing Cr ions [29]. In Fig. 9 we show the PL spectrum of the (CdTe,CdCrTe)/CdMgTe heterostructure containing two CdTe and two CdCrTe QWs with different Cr concentration. Free excitonic emission was observed from each of the QWs. Fig. 9 shows that intensity of QW PL is strongly reduced in QWs containing Cr ions.

There are two possible explanations of such reduction of the PL intensity. Reduction of the PL intensity can be explained by either high efficiency of the bypassing process or by Auger-type energy transfer. In the case of the dominant Auger effect (excitonic emission is quenched by Auger-type energy transfer from exciton to Cr ions) we expected both the reduction of the PL intensity and the shortening of the PL decay time (not expected if the bypassing process dominates). High efficiency of the Auger process could thus be confirmed by PL kinetics studies. PL kinetics was measured under a 2 ps long pulsed excitation [29]. The results of PL kinetics experiments are shown in Fig. 10. We observe a dramatic shortening of the PL decay times from 490 ps (30 ML wide) and 320 ps (6 ML wide) for the CdTe QWs to 200 ps (20 ML wide) and 75 ps (12 MLwide) for the CdCrTe QWs. The shortest PL decay time was observed for the QW with the highest Cr contents. The results of PL kinetics indicate thus that the excitonic Auger effect is efficient chromium-related PL quenching mechanism. The bypassing process, dominant in the bulk samples, is here less important. To get a ‘‘rough’’

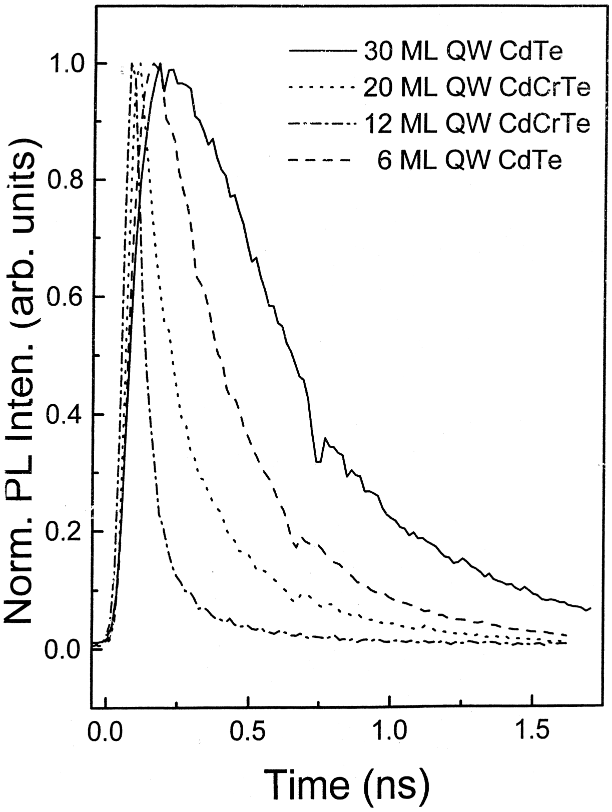

Fig. 10. PL kinetics measured at 2 K for two CdTe and two CdCrTe QWs in the (CdTe,CdCrTe)/CdMgTe heterostructure.

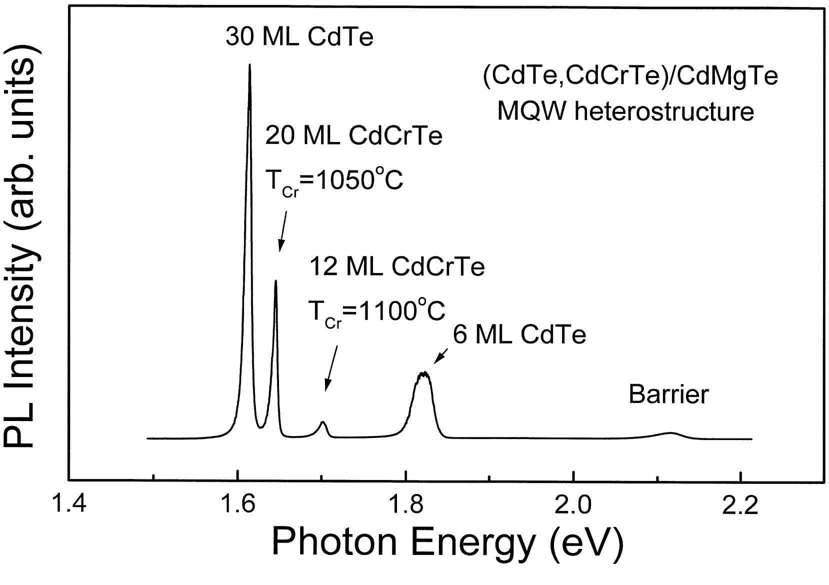

Fig. 9. 2 K PL spectrum of the multiple (CdTe,CdCrTe)/CdMgTe QW structure containing two CdTe QWs (30 monolayer (ML) and 6 ML wide) and two Cr containing CdCrTe QWs (20 ML and 12 ML wide). Chromium fraction in CdCrTe QW increases with an increase of temperature of chromium cell, which is given in the figure.

estimation of the efficiency of the Auger transition we employed Inokuti-Hirayama theory of the resonant energy transfer [29]. Rough estimations of the process indicates that Auger-type mechanism of chromium ionisation is efficient only if one or more of Cr ions is located within the Bohr radius of the exciton.

5. Conclusions

Among various energy transfer processes and processes of indirect excitation of 4f–4f emission of RE ions (3d–3d for TM ions), Auger-type processes are shown to be important. In particular, these processes are shown to be highly efficient for adjacent centres, when RE (TM) ion is closely associated with emission activator. Our recent PL investigations show that Auger-type processes are also efficient in low dimensional quantum well structures.

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