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# Excitons and excitonic molecules in mixed $Zn(P_{1-x}As_x)_2$ crystals

O.A. Yeshchenko\*, M.M. Biliy, Z.Z. Yanchuk

Physics Faculty, Kyiv Taras Shevchenko University, 6 Akademik Glushkov Prosp., 03127 Kyiv, Ukraine

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#### Abstract

Low-temperature (1.8 K) excitonic absorption, reflection and photoluminescence spectra of mixed  $Zn(P_{1-x}As_x)_2$  crystals have been studied at x = 0.01, 0.02, 0.03 and 0.05. Energy gap and rydbergs of excitonic B-, C- and A-series decrease monotonically with increasing of x. The spectral half-widths of the absorption n = 1 lines of the B- and A-series increase monotonically with increase in x due to fluctuations of crystal potential. Emission lines of excitonic molecules have been observed in photoluminescence spectra of  $Zn(P_{1-x}As_x)_2$  crystals. The binding energy of the molecule increases with increase in x due to the decrease of the electron-hole mass ratio. © 2001 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

 $\beta$ -ZnP<sub>2</sub> (further ZnP<sub>2</sub>) and ZnAs<sub>2</sub> crystals are strongly anisotropic direct-gap semiconductors, which are characterized by the same symmetry group  $C_{2h}^5$  (monoclinic syngony). The energy gaps of these crystals are the following: 1.6026 eV for ZnP<sub>2</sub> [1] and 1.052 eV for ZnAs<sub>2</sub> [2,3]. Besides the same symmetry of the lattice, the similarity of ZnP<sub>2</sub> and ZnAs<sub>2</sub> exists in the structure of energy bands and exciton states, namely, three excitonic series are observed in the absorption spectra of

*E-mail address:* yeshchen@phys.univ.kiev.ua (O.A. Yeshchenko).

these crystals: dipole allowed C-series at  $\mathbf{E} || Z(\mathbf{c})$ , forbidden B-series at  $\mathbf{E} \perp Z(\mathbf{c})$  and partially allowed A-series at  $\mathbf{E} \| X$  polarizations (see, e.g. Refs. [1,4–6] for ZnP<sub>2</sub> and Refs. [2,3,7] for ZnAs<sub>2</sub>). In the photoluminescence (PL) spectra of these crystals at  $\mathbf{E} \| Z(\mathbf{c})$  polarization, a series of lines caused by the radiative transitions from the ground n = 1 and excited n = 2, 3 states of allowed S-paraexciton (C-series) is observed (see, e.g. Ref. [5] for  $\beta$ -ZnP<sub>2</sub> and Ref. [2] for ZnAs<sub>2</sub>). Besides this emission series, the so-called B-line is observed in the PL spectra of  $ZnP_2$  at  $E \perp Z(c)$ . B-line occurs due to the radiative transitions from the ground state of forbidden S-orthoexciton and corresponds to  $B_1$ -line of absorption B-series. Serial laws of excitons in ZnP2 are characterized by the following [1]: for C-series, parameters rydberg is

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<sup>\*</sup>Corresponding author. Tel.: + 380-44-266-4587; fax: + 380-44-266-4036.

 $Ry_{C} = 41.3 \text{ meV}$ , short-range correction to the hydrogenlike  $E(n) \propto n^{-2}$  law is  $\Delta_{\rm C} = 1.4$  meV; for A-series  $Ry_A = 41.3 \text{ meV}$ ,  $\Delta_A = 14.1 \text{ meV}$ ; for Bseries  $Ry_B = 45.8 \text{ meV}$ ,  $\Delta_B = 0.^1$  Thus, for C- and B-series the values of corrections are small with respect to rydbergs. Therefore, these series can be fitted with high precision by the law  $E(n) = E_g$  – the following values:  $Ry/n^2$  that gives  $Ry_C = 39.6 \text{ meV}$  and  $Ry_B = 44.0 \text{ meV}.^2$  For Aseries, the correction  $\Delta$  is rather large, and therefore, the fitting of this series by the abovementioned hydrogenlike function gives  $Ry_A = 26.5 \text{ meV}$  that differs appreciably from the above value. In ZnAs<sub>2</sub> crystals, the exciton parameters are the following:  $Ry_C = 12.5 \text{ meV}$ and  $Ry_B = 15.1 \text{ meV}$  [2,3,7]. As to the A-series, an ambiguity of the identification of some lines of this series with concrete values of the quantum number *n* still exists. It is not known whether there are observed lines n = 1, 2, 3 in absorption spectra or only the n = 1, 2 ones [3]. Therefore, we do not give here the value of A-series rydberg in ZnAs<sub>2</sub>.

In ZnP<sub>2</sub> crystals, an excitonic molecule (EM or biexciton) with rather high binding energy  $E_{\text{bex}}^{\text{b}} =$  $6.7 \text{ meV} = 0.15 E_{\text{ex}}^{\text{b}}$  exists, where  $E_{\text{ex}}^{\text{b}}$  is the binding energy of the lowest-energy S-orthoexciton. Proceeding from the theory [8], high relative binding energy of biexciton  $E_{\text{bex}}^{\text{b}}/E_{\text{ex}}^{\text{b}} = 0.15$  in ZnP<sub>2</sub> can be explained by the rather small ratio of effective masses of electron and hole  $\sigma = m_e/m_h = 0.06$ . In ZnP<sub>2</sub>, the Ems manifest themselves in PL spectra due to two-electron and two-photon radiative transitions from the ground state of molecule [9–11]. Two-electron transitions go from the ground state of biexciton to states of excitons of C- and A-series (further C- and A-excitons, correspondingly). The result of such transitions is the appearance of inverse hydrogen-like series of luminescence (further  $M_{\rm C}$ - and  $M_{\rm A}$ -series, correspondingly). The experimental evidences of existence of the biexcitons in ZnAs<sub>2</sub> are absent at

present, though by theoretical estimations they should have binding energy  $\approx 1.6 \text{ meV}$ .

In the present work, we study the mixed crystals (solid solutions) of isovalent substitution  $Zn(P_{1-x}As_x)_2$ , which, as we know, were not studied earlier.  $Zn(P_{1-x}As_x)_2$  belong to the mixed crystals of A<sup>II</sup>B<sup>V</sup> type, which are poorly investigated. At present, the mixed crystals of  $A^{III}B^{V}$ type are the most investigated [12] (review). For the first time, direct exciton transitions in absorption spectra of  $A^{III}B^V$  crystals were observed in  $In_{1-x}Ga_xP$  [13], and also in  $Al_xGa_{1-x}As$  [14]. Later, narrow excitonic peaks n = 1, 2 were observed in absorption spectra of direct-gap  $GaAs_{1-x}P_x$  [15]. Also, excitonic states are rather well investigated in A<sup>II</sup>B<sup>VI</sup> mixed crystals, e.g. in  $CdS_{1-x}Se_x$  [16],  $ZnSe_{1-x}Te_x$  [17], etc. In the present work, the low-temperature (1.8 K) absorption, reflection and photoluminescence spectra of  $Zn(P_{1-x}As_x)_2$  crystals have been studied at small levels of substitution of P by As:  $x \leq 0.05$ . Experimental dependences of parameters of excitons and excitonic molecules on x were obtained.

# 2. Technology of growing of mixed $Zn(P_{1-x}As_x)_2$ crystals

The technological operations of synthesis and growing of ZnP2 crystals were carried out according to Gorban et al. [6]. To obtain  $Zn(P_{1-x}As_x)_2$ crystals, the doping by arsenic was carried out during growth of crystals from a gas phase. Using the previously synthesized and twice purified by resublimation  $ZnP_2$  (weight 8 g) five portions approximately of identical mass were prepared. Each portion was loaded in a quartz ampoule (length—165 mm, bore diameter—18 mm) with a cone-shaped spout. As on air, the arsenic rather promptly oxidizes, for doping we have used the compound of arsenic ZnAs<sub>2</sub>, which at temperature of growth of ZnP2 crystals (940°C) dissociates into Zn and As. The mass of portion of ZnAs<sub>2</sub> was taken so that at given mass of ZnP<sub>2</sub>, contents of arsenic made up a part necessary for us in relation to the contents of phosphorus (in àtomic%). One ampoule was control, i.e. ZnAs<sub>2</sub> was not loaded During pumping out (to there. pressure

<sup>&</sup>lt;sup>1</sup>It should be noted that  $\Delta_{\rm B} = 0$  only for the B-series lines with  $n \ge 2$ . The n = 1 line deviates from the "pure" hydrogenlike law. The value of B-series rydberg Ry<sub>B</sub> = 45.8 meV is obtained from the fitting of the B-series lines with  $n \ge 2$ .

<sup>&</sup>lt;sup>2</sup> This value is obtained from the fitting of all the B-series lines including n = 1.

 $2 \times 10^{-5}$  mm of Hg), the ampoules were heated up to a temperature of 250°C during 1 h and were unsoldered from vacuum installation by the oxygen burner.

#### **3.** Exciton spectra of $Zn(P_{1-x}As_x)_2$ crystals

Absorption, reflection and luminescence spectra of  $Zn(P_{1-x}As_x)_2$  crystals at x = 0.01, 0.02, 0.03 and 0.05 and pure  $ZnP_2$  (x = 0) are presented in Figs. 1–3. The  $Zn(P_{1-x}As_x)_2$  crystals are directgap semiconductors as well as  $ZnP_2$ . One can see from the figures that, in the mixed crystals, the same excitonic C-, B- and A-series are observed, as in pure  $ZnP_2$ . Let us note the doublet structure of an absorption n = 1 line of B-series of a crystal with x = 0.02 (Fig. 1). Proceeding from intensities and half-widths of the components of this doublet, we have made a conclusion, that narrow high-

В,

B₂↓

B₂↓

В

B

0.05

0.03

Α

А

Α,

0.02

0.01

В

В

В

В

В

Absorption kd (arb. un.)

1.54 1.55 1.56 1.57 1.58 1.59 1.60 Photon energy (eV) Fig. 1. Absorption spectra of  $Zn(P_{1-x}As_x)_2$  crystals at temperature 1.8 K. Observation conditions:  $\mathbf{q} \perp (110)$ ,  $\mathbf{E} \perp Z(\mathbf{c})$ —for crystals with x = 0, 0.01, 0.02 and 0.03;  $\mathbf{q} \perp (100)$ ,  $\mathbf{E} \perp Z(\mathbf{c})$ —for crystals with x = 0.05.



Fig. 2. Reflection spectra of  $Zn(P_{1-x}As_x)_2$  crystals at temperature 1.8 K. Observation conditions:  $\mathbf{q} \perp (100)$ ,  $\mathbf{E} \parallel Z(\mathbf{c})$ .

energy component is n = 1 line of B-series. An origin of low-energy component, which is missing for crystals with  $x \neq 0.02$ , is not clear. Let us note, that in contrast to ZnP<sub>2</sub> crystals, where in optical spectra the lines up to n = 7 for B-series and to n = 4 for A- and C-series are observed, in the spectra of  $Zn(P_{1-x}As_x)_2$  crystals of various thicknesses, excitonic lines only with n = 1, 2 are observed. Probably, this fact is a result of "blurring" of the band edges, which takes place owing to fluctuations of crystal potential, caused by chaotic distribution of As atoms in sites of crystal lattice at substitution of P atoms. With increase in concentration x, spectral lines shift to the low-energy side, which is caused by the decrease of energy gap. This shift could be expected, taking into account the fact, that in  $ZnAs_2$ , energy gap is 0.55 eV smaller than in  $ZnP_2$ . The dependence  $E_g(x)$  is given in Fig. 4(a). But besides the trivial decrease of  $E_{\rm g}$  with the increase of x, there is also the decrease of the excitonic



Fig. 3. PL spectra of  $Zn(P_{1-x}As_x)_2$  crystals at temperature 1.8 K.

series rydbergs (see Fig. 4(b)). Values of  $E_g$  and rydbergs were obtained from fitting of excitonic series by simple hydrogenlike dependence:  $E(n) = E_g - Ry/n^2$ .

There were also studied dependences of the halfwidths of absorption n = 1 lines of B- and Aseries. These dependences are given in Fig. 5.<sup>3</sup> It is seen, that half-widths of B<sub>1</sub>- and A<sub>1</sub>-lines increase monotonously with the increase of x. As known, the increase of half-width of exciton lines is the result of fluctuations of crystal potential and respective fluctuations of  $E_g$ . The theory of influence of fluctuations of composition x on half-width of exciton absorption lines was developed in Ablyazov et al. [18], where two extreme cases were considered. The first case takes place, if the effective size of area of the crystal potential fluctuation  $R_D = \hbar/(2MD)^{1/2}$ , where *M* is the total mass of exciton and D(x) = W(x) - W(0) (W(x) is the half-width of exciton line), is much larger than the exciton Bohr radius:  $R_D \ge a_{ex}$ . Such situation, as a rule, takes place, if the effective masses of electron and hole are small and differ slightly:  $m_e \sim m_h$ . In this case, *D* should depend on *x* as

$$D(x) = 0.08 \frac{\alpha^4 M^3 x^2 (1-x)^2}{\hbar^6 N^2},$$
(1)

where  $\alpha = dE_g/dx$ , *N* is the concentration of sites of lattice, where the substituting atoms (As in our case) can "sit". Such a situation occurs in many semiconductors, in particular in A<sup>III</sup>B<sup>V</sup> crystals. The second case takes place at  $R_D \ll a_{ex}$ . It takes place at  $m_h \gg m_e$ . In this case, dependence D(x) has such character

$$D(x) = 0.5\alpha \left(\frac{x(1-x)}{Na_{\rm ex}^3}\right)^{1/2}.$$
 (2)

The experimental dependences D(x) of absorption **B**<sub>1</sub>- and **A**<sub>1</sub>-lines of  $Zn(P_{1-x}As_x)_2$  crystals were fitted by functions (1) and (2). It is seen from Fig. 5, that the experimental points are badly fitted both by functions (1) and (2). It becomes clear if one estimates the effective size of area of fluctuation  $R_{\rm D}$  for different x. Estimations give:  $R_{\rm D} =$ 82A at x = 0.01,  $R_{\rm D} = 62A$  at x = 0.02 and  $R_{\rm D} =$ 30A at x = 0.05. In ZnP<sub>2</sub>, the exciton Bohr radius is  $a_{\text{ex}} = 15A$ . Thus, for  $\text{Zn}(P_{1-x}\text{As}_x)_2$ :  $R_{\text{D}} > a_{\text{ex}}$ , but the condition  $R_{\rm D} \gg a_{\rm ex}$  is not fulfilled, i.e. in studied crystals, the intermediate case takes place which is, nevertheless, more close to Eq. (1). In  $Zn(P_{1-x}As_x)_2$ , the condition  $R_D \gg a_{ex}$  is not fulfilled due to rather large total mass of exciton and appreciable difference of electron and hole masses  $(m_h \gg m_e)$ . Therefore, since the intermediate case takes place, the experimental dependences were fitted by the function

$$D(x) = (1 - c)D_1(x) + cD_2(x),$$
(3)

which is the superposition of functions  $D_1(x)$  of type (1) and  $D_2(x)$  of type (2), c is the weighting factor. It is seen from Fig. 5, that function (3) fits experimental dependences D(x) very well, which confirms our assumption of an intermediate

<sup>&</sup>lt;sup>3</sup>An absence of data on the half-widths of lines for crystal with x = 0.03 is due to the fact that sample with x = 0.03 was rather thick, therefore, full absorption took place in B<sub>1</sub> and A<sub>1</sub> lines. So, it was not possible to determine correctly the half-widths of these lines. As we have pointed out above, half-width of B<sub>1</sub> line for crystal with x = 0.02 was determined for the high-energy component of doublet.



Fig. 4. Dependences of energy gap (a) and rydbergs of excitonic series (b) in  $Zn(P_{1-x}As_x)_2$  crystals on level of substitution x.



Fig. 5. Dependences of spectral half-widths of absorption n = 1 lines of excitonic B- and A-series of  $Zn(P_{1-x}As_x)_2$  crystals on level of substitution *x*.

case. At known  $\alpha = dE_g/dx$ , *M* and  $a_{ex}$ , the fitting of D(x) by function (3) gives the possibility to estimate *N* and *c*. The fitting of dependences D(x)for absorption n = 1 lines of B- and A-series gives:  $N_{\rm B} = N_{\rm A} = 0.026A^{-3}$ ,  $c_{\rm B} = 0.05$  and  $c_{\rm A} = 0.32$ . Concentration of sites of the lattice, where the substituting atoms can "sit", can be estimated as  $N \sim a_{\rm P-P}^{-3} = 0.09A^{-3}$ , where  $a_{\rm P-P} = 2.20A$  is the length of P–P bond in ZnP<sub>2</sub> crystal. Thus, taking into account estimation character of value  $N \sim 0.09A^3$ , the values obtained from fitting and estimated from the length of P-P bond are in quite good agreement. The fact, that  $c_A$  is considerably larger than  $c_B$ , indicates that in dependence D(x) for n = 1 line of A-series the function  $D_2(x)$  of type (2) makes a considerably larger contribution than for n = 1 line of B-series. It is clear that since A-exciton has considerably smaller binding energy than B-exciton, it has larger Bohr radius. This makes A-exciton closer to case (2) when compared with B-exciton.

#### 4. Excitonic molecules in $Zn(P_{1-x}As_x)_2$ crystals

In PL spectra of mixed  $Zn(P_{1-x}As_x)_2$  crystals, as well as  $ZnP_2$  crystals, there are  $M_C$  and  $M_A$  lines (Fig. 3). These lines are due to radiative transitions from the ground state of excitonic molecule to the ground state of C-exciton ( $M_C$ -line) and ground state of A-exciton ( $M_A$ -line). The EMs are rather well investigated in pure  $ZnP_2$ . As well as in pure  $ZnP_2$ , the M-lines demonstrate square-law character of dependence of their intensity on excitation intensity that confirms their biexcitonic nature. Binding energy of EM can be determined from PL spectrum as  $E_{bex}^b = 2E_{ex} - E_{C1} - \hbar\omega_{M_C}$ , where  $E_{ex}$ 



Fig. 6. Dependence of binding energy of excitonic molecule in  $Zn(P_{1-x}As_x)_2$  crystals on level of substitution x. Binding energy is presented in meV (a), and in units of a binding energy of B-exciton (b).

is the energy of the lowest B-exciton ground state (B<sub>1</sub>-line),  $E_{C1}$  is the energy of the ground state of allowed C-exciton, and  $\hbar\omega_{M_C}$  is the energy of a photon of  $M_{\rm C}$ -line. The obtained dependence of binding energy of biexciton on x is presented in Fig. 6. It is seen, that contrary to rydbergs of excitonic series, the binding energy of EM increases with the increase of x both by absolute value and in relation to binding energy of the lowest B-exciton. Such behaviour of  $E_{bex}^{b}$  with the increase of x can be explained. As known (e.g. from Ref. [8]), the ratio  $E_{\text{bex}}^{\text{b}}/E_{\text{ex}}^{\text{b}}$  is a function of the ratio of effective electron and hole masses  $\sigma = m_{\rm e}/m_{\rm h}$ , and with the decrease of  $\sigma$ , the ratio of binding energies of biexciton and exciton increases. At substitution of atoms of one type by atoms of another type (in our case of P atoms by As atoms), the variation of parameters of energy band structure occurs, in particular the variation of effective masses of carriers. Since dielectric constant has close values in ZnP2 and ZnAs2 crystals,  $\varepsilon$  should not vary considerably in mixed crystals. Therefore, the decrease of exciton rydbergs with the increase of x is an evidence of the decrease of reduced effective mass of carriers. Reduced mass  $\mu = m_e m_h / (m_e + m_h) = m_e / (1 + \sigma)$ at small  $\sigma < 0.1$  is  $\mu \approx m_e$ . Therefore, the decrease of

 $\mu(x)$  reflects the decrease of  $m_e(x)$ . Such a behaviour of  $m_e(x)$ , i.e. the decrease of effective electron mass with the decrease of a direct energy gap, is well known, e.g. for  $A^{III}B^V$  semiconductors [12]. The decrease of  $m_e(x)$  can explain the decrease of  $\sigma$ , and so the respective increase of binding energy of biexciton. But how does effective mass of a hole behave? The dependence  $m_h(x)$  can be estimated. Knowing the ratio of the reduced effective masses  $\mu(x)/\mu(0) = Ry(x)/Ry(0)$  (this equality is carried out at  $\varepsilon = \text{const}$ ) and the ratio  $\sigma(x)/\sigma(0)$ ,<sup>4</sup> one can calculate the ratios  $m_e(x) \times /m_e(0)$  and  $m_h(x)/m_h(0)$  according to the following expressions:

$$\frac{m_{\rm e}(x)}{m_{\rm e}(0)} = \frac{\mu(x)}{\mu(0)} \frac{1 + \sigma(x)}{1 + \sigma(0)},$$
$$\frac{m_{\rm h}(x)}{m_{\rm h}(0)} = \frac{m_{\rm e}(x)}{m_{\rm e}(0)} \frac{\sigma(0)}{\sigma(x)}.$$
(4)

Results of respective estimations are given in Fig. 7. It is seen, that with the increase of x, the

<sup>&</sup>lt;sup>4</sup> For ZnP<sub>2</sub>,  $\sigma = 0.06$ . For Zn(P<sub>1-x</sub>As<sub>x</sub>)<sub>2</sub>, the values of  $\sigma(x)$  can be estimated from the dependence  $[E_{bex}^b/E_{ex}^b](\sigma)$  [8] and the experimental dependence  $[E_{bex}^b/E_{ex}^b](x)$ . Respective estimations give:  $\sigma(0.01) = 0.051$ ,  $\sigma(0.02) = 0.045$ ,  $\sigma(0.03) = 0.037$  and  $\sigma(0.05) = 0.026$ .



Fig. 7. Calculated dependences of effective masses of electron and hole in  $Zn(P_{1-x}As_x)_2$  crystals on level of substitution *x*.  $m_{e,h}(0)$  are the masses of carriers in  $ZnP_2$  crystal (x = 0).

electron mass decreases, and the hole mass increases. Thus, the increase of  $m_h$  occurs even faster than the decrease of  $m_e$ . This fact becomes clear if one takes into account, that in ZnP<sub>2</sub>, the conduction band originates mainly from Zn ions and the valence band—from P ions [19]. Therefore, substitution of P atoms by As atoms should first of all have an influence on parameters of the valence band, in particular, on the hole effective mass. Thus, simultaneous decrease of  $m_e$  and increase of  $m_h$  result in the appreciable decrease of ratio  $\sigma = m_e/m_h$ , and therefore, in the respective increase of binding energy of the EM.

# 5. Conclusions

In conclusion, we point out the following obtained results. In mixed  $Zn(P_{1-x}As_x)_2$  crystals at small x in optical spectra the same as in  $ZnP_2$  three excitonic C-, B-, and A-series are observed.

With the increase of x, the energy gap and the excitonic series rydbergs decrease. With the increase of x, the spectral half-widths of excitonic absorption lines increase, that is result of increasing role of the fluctuations of composition and, respectively, the fluctuations of crystal potential. In PL spectra, the lines of excitonic molecules are observed. It is revealed, that with the increase of x, the binding energy of EM increases. The increase of binding energy of biexciton occurs owing to the decrease of the ratio of effective masses of electron and hole, that is result of the simultaneous decrease of electron mass and increase of hole mass.

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