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OPTICAL PUMPING MEASUREMENT OF VALENCE BAND SPIN-ORBIT SPLITTING IN Ga_xIn_{1-x}As_yP_{1-y}

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We have studied by optical pumping the band structure of $Ga_xIn_{1-x}As_yP_{1-y}$ which can be grown nearly lattice-matched on InP ($x \simeq 0.47y$) and has important opto-electronic device applications. We have determined the band gaps $(E_0, E_0 + A_0)$ and the light hole mass as a function of composition. We explain the measured variation of these parameters by an extension of the ternary alloys band theory.

I. Principle of the Experiment

Optical pumping, i.e. excitation of the semiconductor by circularly polarized σ^{\pm} light of energy $h\nu$ larger than the band gap E_0 , creates conduction electrons with a polarization P_i characteristic of the band structure and of the light polarization [1]. Measuring the variation of P_i as a function of $h\nu$ gives informations on the semiconductor band structure [2].

In a p-type zinc-blende direct gap semiconductor P_i is equal to +0.5 for σ^- excitation of energy $h\nu = E_0$ [1]. When $h\nu$ is increased from E_0 to $E_0 + \Delta_0$ (where Δ_0 is the valence band spin-orbit splitting) there is an additional contribution of opposite sign to P_i due to optical transitions from the split-off valence band. This reduces P_i , and a kink results in the variation of P_i with $h\nu$ [2] at $h\nu = E_0 + \Delta_0$ as was first evidenced in pure GaAs [3][4]. The detailed shape of $P_i(h\nu)$ is determined by the joint density of states relevant to the optical transitions from the valence bands to the conduction band. It can thus be related to the carriers effective masses by Kane's 4-band \vec{k} , \vec{p} perturbation theory through the parameter β [2]

$$\beta = \frac{4}{3} \frac{(1/m_c) + 0.75(1/m_\ell) + 0.25(1/m_h)}{(1/m_\ell) - (1/m_h)}, \qquad (1)$$

where ${\tt m}_{\rm C}$ is the electron effective mass, ${\tt m}_{\rm l}({\tt m}_{\rm h})$ the light (heavy) hole mass.

To deduce $P_i(h\nu)$ from the luminescence circular polarization we observe the band-to-shallow-acceptor luminescence line, due to thermalized electrons at the bottom of the conduction band. For σ^- excitation, this line polarization is equal to 0.5P(h\nu) [1]. The steadystate electronic polarization P(h\nu) is the product of $P_i(h\nu)$ by a relaxation term :

$$P(hv) = P_{\tau}(hv) T_{\tau}/(T_{\tau}+\tau)$$
(2)

where \textbf{T}_{1} is the electron spin relaxation time, τ its lifetime.

Consequently, by measuring the luminescence circular polarization, it is possible to deduce the variation of $P_1(h\nu)$ through eq.(2), provided the ratio $T_1/(T_1+\tau)$ is energy independent. This restriction sets conditions on the electron thermalization in the conduction band [4] [5].

II. Experiment and Analysis of the Results

We have applied the above technique to liquid phase epitaxy layers of Zn p-doped $Ga_xIn_{1-x}As_yP_{1-y}$ of concentration ~ $10^{24}m^{-3}$. The sample compositions and the experimental results are summarized in Table I. The samples cooled at 1.7K or 77K are excited by circularly polarized light from an infrared c.w. dye laser [6] (for y = 0.34) or a mercury lamp filtered by a monochromator (y>0.34). We observe their luminescence and polarized luminescence as a function of excitation energy hv.

Table I

| x | У | E _O eV | E _o +Δ _o eV | β | m _c /m _o | m _l /m _o |
|------|------|----------------------------|--------------------------------------|---------|--------------------------------|--------------------------------|
| 0 | 0 | 1.423 ±0.0005 ^a | 1.531±0.0025 ^a | 4.1±0.5 | 0.079 ^a | 0.12 ±0.01 ^a |
| 0.16 | 0.34 | 1.19 ±0.01 | 1.37 ±0.005* | 4.2±0.4 | 0.066 ^b | 0.109±0.011 |
| 0.30 | 0.61 | 1.035 ±0.01 | 1.29 ±0.01 | 3.4±0.4 | 0.052 ^b | 0.072±0.01 |
| 0.39 | 0.84 | 0.875 ±0.01 | 1.18 ±0.01 | 3.2±0.3 | 0.046 ^b | 0.061±0.007 |
| 0.47 | 1 | 0.80 ±0.01 | 1.15 ±0.01* | 3.0±0.2 | 0.041 ^b | 0.051±0.005 |

The data are taken at 1.7K and m_0 is the free electron mass: $a_{\text{Ref}}[17]$

^bRef.[7]

*Calculated from the data of [17] *These results were the same at 1.7K and 77K



Fig.l Variation of the electronic polarization P with excitation energy $h\nu$: The dots are the experimental data, the solid line is calculated from [2] The dots on Fig.1 present the experimental variation of the electronic polarization $P(h\nu)$ obtained at 77K on a sample with x = 0.16, y = 0.34, the luminescence of which is centered at 1.170 eV. These data are taken at constant low excitation level, necessary to keep the relaxation factor constant in eq.(2). The full curve is the variation of P;(hv) predicted by D'yakonov and Perel' [2]. The polarization is normalized to the value at the band gap P = 0.39. The kink occurs at 1.370 ± 0.005 eV, which yields $E_0 + \Delta_0$. The energies are scaled using for Eo the threshold of the luminescence excitation spectrum (E = 1.19

 ± 0.01 eV in this sample). We deduce $\Delta_0 = 0.18 \pm 0.015$ eV. From the shape of the curve we find $\beta = 4.2 \pm 0.4$. The good fit of these data

shows a posteriori that the relaxation factor of eq.(2) is independent of hv.

The other data are given in Table I. In the samples studied at 1.7K and 77K, $E_0+\Delta_0$ takes the same value for both temperatures. For alloys with y > 0.34 the resolution on $E_0+\Delta_0$ is only 0.01 eV, because of weaker signal due to mercury lamp excitation.

III. Discussion

The dependence of Pi(hv) on β given in [2] neglects the admixture of the upper conduction band Γ_5 into Γ_5 in a non-centrosymmetric semiconductor, as well as the change with energy of the conduction band wavefunction. However due to the experimental uncertainty our results on β are not affected by these approximations. We observe a decrease in β with increasing y. The conduction mass m_c has been measured in $Ga_x In_{1-x} As_y P_{1-y}$ [7][8]. The heavy hole mass m_h is close to 0.5 in InP, InAs and GaAs [9]. Moreover measurements of m_h give about 0.5 for two different compositions of this quaternary compound [10]. So we assume that m_h keeps this same value for all alloys. Using eq.(1) and the measurements of m_c [7] we deduce the light hole masses as a function of composition. These values are close to linear interpolation between those in InP, InAs and GaAs, and are in agreement with magnetoreflectance measurements [10].



Fig.2 Variation of $E_0(\mathbf{X})$ and $E_0 + \Delta_0(\bullet)$ as a function of alloy composition in $Ga_X In_{1-x} As_y P_{1-y}$: The data of [11] are shown by triangles. The solid lines are our predictions

The variations with composition of E_0 and $E_0+\Delta_0$ are plotted on Fig.2. The previous determinations of E_0 and $E_0+\Delta_0$ in Ga0.21In0.79As0.54P0.46, shown by triangles, fall close to our results [11]. The very recent room temperature electroreflectance measurements of E_0 and $E_0+\Delta_0$ [12] indicate a more pronounced bowing than our results. Our value of Δ_0 in Ga0.47In0.53As agrees with the room temperature determination of [13]. The theoretical curve is calculated using the dielectric model of Van Vechten and Bergstresser [14]: as in a ternary alloy, we separate the E_0 bowing into the virtual crystal (VC) and disorder contributions.

Since the VC term is related to lattice constant variation [15], it is negligible in $Ga_XIn_{1-x}As_yP_{1-y}$, as all alloys are lattice matched to InP. Here the disorder term includes contributions from anion and cation. We take it equal to :

$$(1/A) \left[C^{2}_{As-P} y(1-y) + C^{2}_{Ga-In} x(1-x) \right], \qquad (3)$$

where A = 1.0 eV, and C_{AS-P} and C_{Ga-In} are electronegativities differences [14], so that the quadratic term is ~ $0.15y^2$. This term reduces E_0 from the linear interpolation between the InP and virtual crystal $Ga_{0.47}In_{0.53}As$ gaps. The variation with y of E_0+A_0 is predicted following [13] and reducing again the bowing to the disorder contribution (3). The fit is good for both E_0 and E_0+A_0 , whereas the previous procedure gave a systematically too strong bowing for $E_0(y)$ [16]. Our predicted E_0 bowing falls close to the room temperature photoluminescence results on $E_0(y)$ [16].

In conclusion, we have been able to justify the variation of the band gaps in $Ga_xIn_{1-x}As_yP_{1-y}$ by an elementary extension of the ternary alloys theories. The weak experimental bowing comes from the absence of VC contribution in this special case.

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