Optical anisotropy and spin polarization in ordered GaInP

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Optical anisotropy and spin polarization in ordered GaInP

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Spontaneous CuPt-like ordering of $Ga_xIn_{1-x}P$ causes a splitting at the valence band maximum (VBM) and induces an anisotropy in the intensities of the transitions between these split VBM components and the conduction band minimum. We calculate these intensities as function of ordering parameter η, and show that the transition intensities depend strongly on the light polarization $\hat{e}$ and the degree of long-range order η in the sample. Furthermore, for sufficiently

widely observed in vapor phase growth on GaAs (001)

monolayer planes $Ga_{x+\eta/2}In_{1-x-(\eta/2)}$ and $Ga_{x-\eta/2}In_{1-x+\eta/2}$ stacked along the $[\bar{1}11]$ or the $[1\bar{1}1]$ directions (known as the

cessive planes of pure Ga followed by pure In, etc. This type

tive to the random alloy. These effects have been observed in

larized piezoreflectance.[7] However, quantitative analysis of

tion rates between the split VBM components and the conduction band minimum (Γ_{1c}), and the ordering of the band gaps (for

of long-range order. We provide here an easy to use formalism to calculate the intensities at the Γ point as a function of

range order η. Furthermore, we predict that for a sufficiently

Neglecting mixing between valence and conduction

the 6×6 Hamiltonian:

$$H_v=\frac{1}{3}\begin{pmatrix} 0 & \Delta^O-i\Delta^{SO} & \Delta^O & 0 & 0 & \Delta^{SO} \\ \Delta^O+i\Delta^{SO} & 0 & -\Delta^O & 0 & 0 & -i\Delta^{SO} \\ \Delta^O & -\Delta^O & 0 & -\Delta^{SO} & i\Delta^{SO} & 0 \\ 0 & 0 & -\Delta^{SO} & 0 & \Delta^O+i\Delta^{SO} & \Delta^O \\ 0 & 0 & -i\Delta^{SO} & \Delta^O-i\Delta^{SO} & 0 & -\Delta^O \\ \Delta^{SO} & i\Delta^{SO} & 0 & \Delta^O & -\Delta^O & 0 \end{pmatrix}. \tag{1}$$

In this representation all four 3×3 blocks contain the spin-orbit elements, while the two diagonal 3×3 blocks also con-

For the random alloy ($\eta=0$) E_2 and E_3 are degenerate. Thus ordering splits the Γ_{4v} VBM of the random alloy into the

matched to the substrate, an additional strain term, neglected here, has to be added to the diagonal blocks.[8] Here, $\Delta^O(\eta)=c^2\Delta^O(1)$ is the crystal field splitting due to ordering[2] and Δ^{SO} is the spin-orbit splitting. For $Ga_{0.5}In_{0.5}P$, we calculated[2] $\Delta^O(\eta=1)=0.20$ eV and $\Delta^{SO}=0.10$ eV. The eigenvalues and eigenvectors of the valence states can be obtained by solving Eq. (1). The three spin-degenerate energy levels (in decreasing order) $E_1(\Gamma_{4v,5v})$, $E_2(\Gamma_{6v})$, and $E_3(\Gamma_{6v})$ {shifted by $\frac{1}{6}[\Delta^{SO}+\Delta^O(\eta)]$} are

$$E_1(\eta)=\tfrac{1}{2}[\Delta^{SO}+\Delta^O(\eta)],$$

$$E_2(\eta)=+\tfrac{1}{2}\sqrt{[\Delta^{SO}+\Delta^O(\eta)]^2-\tfrac{8}{3}\Delta^{SO}\Delta^O(\eta)}, \tag{2}$$

$$E_3(\eta)=-\tfrac{1}{2}\sqrt{[\Delta^{SO}+\Delta^O(\eta)]^2-\tfrac{8}{3}\Delta^{SO}\Delta^O(\eta)}$$

The valence eigenstates Ψ_v of Eq. (1) are linear combination of the six basis functions $\{p_\nu\hat{\sigma}\}$, where $p_\nu=p_x$, p_y, or p_z are the $L=1$ orbital components and σ are the spinors parallel or antiparallel to the z direction. The conduction states are taken here as $\Psi_c=s\hat{\sigma}$, where s is the $L=0$ orbital component. The transition intensity between Ψ_c and Ψ_v is proportional by the matrix element squared $I_{v,c}=|\langle\bar{\phi}_c|H_{int}|\bar{\phi}_v\rangle|^2\ |\langle\Psi_c|H_{\rm int}|\Psi_v\rangle|^2$, where $H_{\rm int}$ is the interacting Hamiltonian. For *linearly* polarized light along the $[l, m, n]$ direction we have $H_{\rm int}\propto lx+my+nz$, while for *circularly* polarized light $\sigma^\pm$ with angular momentum parallel and antiparallel to z' we have[9] $H_{\rm int}\propto x'\pm iy'$. In general

nation $\sigma^{\pm,0}$ $x'=x$, y, or z. The transition matrix elements

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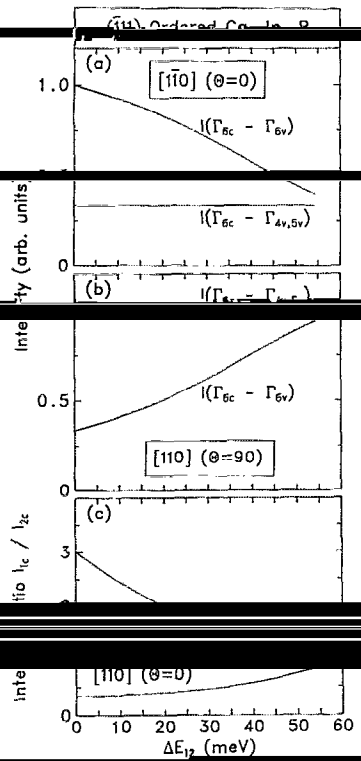

and (b) light polarized alone [110]. Part (c) gives the intensity ratio $I(\Gamma_{6c}-\Gamma_{4v,5v})/I(\Gamma_{6c}-\Gamma_{6v})$.

can be calculated by writing the orbital wave functions and H_{int} in terms of the spherical harmonics Y_{lm} and by noticing that the allowed dipole transitions are for $\Delta m=+1$. This gives the simple selection rule

$$\langle s\hat{\sigma}|\hat{\chi}_{\alpha}|p_{\alpha}\hat{\sigma}'\rangle = c\,\delta_{\alpha,\alpha'}\delta_{\sigma,\sigma'}, \tag{3}$$

where c is a normalization parameter.

We have calculated the transition intensities $I_{v,c}$ between the valences $|1\rangle$ and $|2\rangle$ and the conduction state in ($\bar{1}11$) ordered $Ga_{0.5}In_{0.5}P$ as a function of $\Delta E_{12}(\eta)=E_1(\eta)-E_2(\eta)$. We consider linearly polarized light with polarization in between. Figure 1 shows the calculated intensities for the transitions $\Gamma_{4v,5v}-\Gamma_{6c}$ and $\Gamma_{6v}-\Gamma_{6c}$ as function of ΔE_{12} for polarization $\hat{e}\|[1\bar{1}0]$ (part a) and $\hat{e}\|[110]$ (part b). Measured intensities include, in addition to $I_{v,c}$, line broadening, joint density of states, and the Boltzmann population factors. We see from Figs. 1(a) and 1(b) that the intensity $I(\Gamma_{6c}-\Gamma_{4v,5v})$ does not depend on ΔE_{12}, thus on degree of ordering. This is so since there is no coupling between $\Gamma_{4v,5v}$ and the other

depends strongly on ΔE_{12} because the coupling between the

We will distinguish in what follows two limits of cou-

$$\frac{I(\Gamma_{6c}-\Gamma_{4v,5v})}{I(\Gamma_{6c}-\Gamma_{6v})}=\begin{cases}1/3; & \hat{e}\|[1\bar{1}0]\\ 3; & \hat{e}\|[110]\end{cases} \tag{4}$$

Since in this "weak ordering" regime, $\Gamma_{4v,5v}$ and Γ_{6v} are nearly degenerate, only the combined intensity $I(\Gamma_{6c}-\Gamma_{4v,5v})+I(\Gamma_{6c}-\Gamma_{6v})$ can be measured. For $\Delta E_{12}=0$ this combined intensity is the same for the [110] or the [1$\bar{1}$0] polarization [Figs. 1(a) and 1(b)], as appropriate for a cubic system. Thus no polarization dependence should be detected if the two transitions are not resolved. As the degree of order (thus, ΔE_{12}) increase, the intensity ratios $I(\Gamma_{6c}-\Gamma_{4v,5v})/I(\Gamma_{6c}-\Gamma_{6v})$ approaches unity for both polarization directions. In the extreme trigonal limit [Fig. 1(c)]

$$\frac{I(\Gamma_{6c}-\Gamma_{4v,5v})}{I(\Gamma_{6c}-\Gamma_{6v})}=\begin{cases}1; & \hat{e}\|[1\bar{1}0]\\ 1; & \hat{e}\|[110]\end{cases} \tag{5}$$

and the polarization dependence for both transitions $I(\Gamma_{6c}-\Gamma_{4v,5v})$ and $I(\Gamma_{6c}-\Gamma_{6v})$ are given by [Figs. 1(a) and

$$\frac{I_{110}}{I_{1\bar{1}0}}=3. \tag{6}$$

of the polarization angle Θ. We find that in this

$$I(\Theta)=I_{110}\sin^2\Theta+I_{1\bar{1}0}\cos^2\Theta. \tag{7}$$

For the $\Gamma_{6c}-\Gamma_{4v,5v}$ transition [Fig. 2(a)] the intensity is independent of η. For the $\Gamma_{6c}-\Gamma_{6v}$ transition, however, we see a strong dependence on the ordering parameter η. $I(\Theta)$ ing function (at small η) of the polarization angle Θ. Figure larized electroreflectance data of Kanata *et al.*[5] (solid dots). We find that the best fit to the measured intensities is obtained using $\eta=0.58$. Inserting this value into Eq. (2) gives a valence band splitting of $\Delta E_{12}=34$ meV. The directly measured[5] valence band splitting of this sample is

ergetic analysis. The analysis of Ref. 5 of the same data neglected polarization cross-terms[10] and is thus incomplete.

Our model has an interesting conclusion on $I_{v,c}$ vs η: in most previous analyses of experimental data,[6,7,5,11] the intensity ratio of the quasicubic limit [Eq. (4)] was applied to *all* degrees of ordering η. The ordering dependence of the intensity ratio was thus missed. As we see in Figs. 1(c) and

angular momentum along the ordering direction $z'=[111]$.

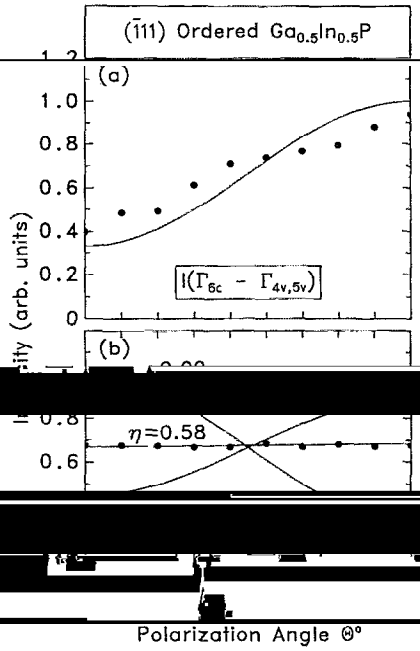

FIG. 2. Calculated transition intensity of $(\bar{1}11)$ ordered $Ga_{0.5}In_{0.5}P$ as a function of polarization angle Θ. Here, $\Theta=0$ denotes light polarized along $[1\bar{1}0]$, while $\Theta=90$ denotes light polarized along $[110]$. (a) The Γ_{6c} $\Gamma_{4v,5v}$ [illegible] at $\eta=0$, 0.58, and 0.87. The corresponding ΔE_{12} are 0, 34, and 50 meV, respectively. The solid dots in (a) and (b) are the experimental data of

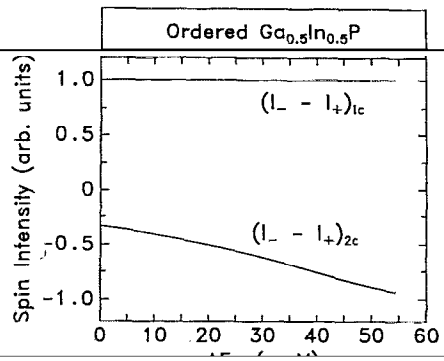

FIG. 3. Calculated spin intensity $(I_- - I_+)_{1c}$ and $(I_- - I_+)_{2c}$ (in arbitrary units) of ordered $Ga_{0.5}In_{0.5}P$ as a function of the valence band splitting ΔE_{12}.

ting than (001) ordered samples, we suggest that (111) ordered III-V alloys (e.g., $Ga_{0.5}In_{0.5}P$) could be a better candidate of spin polarized photoelectron source.

Note that despite the identical optical response with respect to the linearly polarized light along [110] and $[1\bar{1}0]$ of the two CuPt$_B$ [$(1\bar{1}1)$ and $(\bar{1}11)$] subvariants, their response

Using the σ^- light noted above but for (111) ordering, we

top $\Gamma_{4v,5v}$ state is only 20% and the total intensity $I_- + I_+$ is reduced to 55.56% of the intensity for $(\bar{1}11)$ ordering. This difference can be used to distinguish $(1\bar{1}1)$ ordering from $(\bar{1}11)$ ordering, which is not possible using the linearly polarized light. This also indicates that in order to obtain the highest efficiency in generating spin polarized electrons

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$$\uparrow' = \cos\frac{\theta}{2}e^{-i(\varphi/2)}\uparrow + \sin\frac{\theta}{2}e^{i(\varphi/2)}\downarrow,$$

$$\theta \qquad\qquad \theta$$

the spinors parallel and antiparallel to the [001] direction, respectively. The electron spin polarization P is defined as[9,12]

$$P=\frac{I_- - I_+}{I_- + I_+}, \tag{9}$$

where I_- and I_+ are the transition intensities for $\downarrow'$ spin and $\uparrow'$ spin, respectively. In Fig. 3 we show the calculated spin intensities $(I_- - I_+)_{1c}$ and $(I_- - I_+)_{2c}$ for ordered $Ga_{0.5}In_{0.5}P$ alloys. For the random alloy ($\Delta E_{12}=0$) the states $|1\rangle$ and $|2\rangle$ (both Γ_{8v}) are degenerate. In this case, we find that optical pumping from both states leads to a $P=50\%$ electron spin polarization.[9] For ordered alloy ($\eta\neq 0$), the $|1\rangle=\Gamma_{4v,5v}$ and $|2\rangle=\Gamma_{6v}$ states split. We find that photoelectrons generated from the $\Gamma_{4v,5v}$ and the Γ_{6v} states are both fully polarized $P=+100\%$. Hence, if the splitting ΔE_{12} is large enough to allow optical pumping only from the highest $\Gamma_{4v,5v}$ state, the generated photoelectrons can be 100% spin polarized. A theoretical 100% spin polarization of electron can also be achieved by using (001) ordered material [e.g., $Al_{0.5}Ga_{0.5}As$ (Ref. 13)], as proposed by Ciccacci *et al.*[12] However, since $(\bar{1}11)$ ordered material has much larger valence band split-

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$$|\langle\Psi_c|x+y|\Psi_v\rangle|^2=|\langle\Psi_c|x|\Psi_v\rangle|^2+|\langle\Psi_c|y|\Psi_v\rangle|^2.$$

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