Role of symmetry reduction in the polarization dependence of the optical absorption in non-common-atom superlattices

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Ab initio calculations of the dielectric tensor elements show that the mechanism underlying the experimentally observed giant polarization anisotropy of the optical absorption in non-common-atom superlattices is a ''selection rule'' by which the radiation polarization along the 110 or −110 directions *chooses* only one transition of the doublet degenerate in common-atom superlattices. The magnitude of the anisotropy depends *only* on the energy splitting between the two hole states. Our results suggest ways to enhance the anisotropy through manipulation of interface structure or chemistry or strain condition. [S0163-1829(98)51828-0]

It has been found recently[1–3] that a considerable difference exists between the optical properties of the ''non-common-atom'' (NCA) superlattices (SL's), such as (InGa)As-InP or InAs-AlSb, in which the host materials do not share common atoms and the ''common-atom'' (CA) SL's, such as (AlGa)As-GaAs. The optical transmission spectra of InGaAs-InP SL's grown along the [0,0,1] direction display a strong anisotropy of the optical absorption with respect to the angle θ between the photon polarization and the sample x axis. These effects are studied mainly over a 70 meV broad spectral range just above the absorption edge where it has been shown that the sample absorption is stronger when the photon polarization is along the [1,1,0] direction than when it is along the [−1,1,0] direction for light propagating along the SL growth direction. Conversely, CA SL's do not show any polarization anisotropy (PA) of the optical absorption. The main feature distinguishing NCA SL's, let us say, (InGa)As-InP, from the corresponding CA ones [for example, an ideal virtual crystal (InGa)(AsP)-In(AsP)] is the reduction of the crystal field (CF) symmetry, which in the NCA SL's has only 4-point group operations (C_{2v} group) instead of the 8-symmetry operations of the D_{2d} point group of the CA SL's. The C_{2v} point group misses all the symmetry operations related to the z-axis inversion. The symmetry reduction in the NCA SL's stems from the inequivalence of the interfaces between the two C1-A1 and C2-A2 host materials (C and A indicate cation and anion respectively). At a C1-A1/C2-A2 interface, if A1 is the interface plane, the C1-A1 bonds lie in the {−1,1,0} plane, while the C2-A1 bonds (which do not belong to either the host compounds) lie in the {1,1,0} plane. In the CA SL's this situation is compensated at the other interface with the C2-A1 bonds in the {−1,1,0} plane and the C1-A1 bonds in the {1,1,0} plane. This compensation does not occur in NCA SL's because the chemical species forming the bonds themselves are different: there are C1-A1 and C2-A1 bonds at one interface and C2-A2/C1-A2 bonds at the other interface. Consequently, the crystal potential is no longer symmetric with respect to the midpoint of the (InGa)As layer for all the roto-inversion operations of the D_{2d} point group changing z into $-z$, which, in Schönflies notation, are indicated as C_{2x},

C_{2y}, IC_{4z}^{-1}, and IC_{4z}. The symmetry reduction plays a role in the PA of the optical absorption of the NCA SL's. This point has been investigated through envelope function[1] and semiempirical tight-binding[3] approaches. In particular the PA is a forbidden result in the usual $\mathbf{k} \cdot \mathbf{p}$ theory, in which potential fluctuations on the scale of atomic spacing are not taken explicitly into account. However, Krebs and Voisin[1] have shown that the *ad hoc* inclusion of the local symmetry properties in the theoretical description leads to the mixing of the heavy- and light-hole states at the zone center and ultimately to a polarization dependence of the optical absorption coefficient. Similarly, a semiempirical tight-binding calculation,[3] which includes strain effects at the interfaces but does not take into account a possible SL valence-band offset asymmetry,[4] finds a partial admixture of the light and heavy characters in the hole wave functions. This calculation finds that, due to the mixing of the light- and heavy-hole characters, the optical transitions between the three upper valence states and the first conduction state at the zone center are in-plane polarized. This leads to a polarization angle dependence of the oscillator strengths and, ultimately, to a polarization dependence of the absorption spectrum. The theoretical results based on semiempirical approaches find the polarization rate:

$$p(\omega) = (\alpha_{110} - \alpha_{\bar{1}10})/(\alpha_{110} + \alpha_{\bar{1}10}) \tag{1}$$

in qualitative agreement with the experiment but consistently smaller.

In this paper we study how the symmetry properties of the dielectric tensor (DT) determine the conditions leading to the observed in-plane PA and subsequently calculate these effects by using first-principles self-consistent approaches. The aim of our *ab initio* calculation is to assess separately the different effects contributing to the PA of the optical absorption without having to put *ad hoc* parameters into the calculation. In fact, the properties of the crystal potential and the possible band offset noncommutativity are natural outputs of the calculation and therefore the link between the microscopical configuration of the SL interfaces and the properties of the DT is better clarified.

TABLE I. Dielectric tensor elements for CA and NCA SL's. Here $K = 8\pi^2 e^2/\omega^2 m^2 V$. δ remains the same for $\delta(E_c(k) - E_v(k) - \hbar\omega)$. The summation over k is intended only over the IBZ. $\epsilon_{xz} = \epsilon_{zx}$ and $\epsilon_{yz} = \epsilon_{zy}$ are zero.

Dielectric tensor elements	CA (D_{2d})	NCA (C_{2v})
$\epsilon_{xx}(\omega) = \epsilon_{yy}(\omega)$	$K\Sigma_{v,c}\Sigma_k 4(P_x^* P_x + P_y^* P_y)\delta$	$K\Sigma_{v,c}\Sigma_k 2(P_x^* P_x + P_y^* P_y)\delta$
$\epsilon_{zz}(\omega)$	$K\Sigma_{v,c}\Sigma_k 8 P_z^* P_z \delta$	$K\Sigma_{v,c}\Sigma_k 4 P_z^* P_z \delta$
$\epsilon_{xy}(\omega) = \epsilon_{yx}(\omega)$	0	$K\Sigma_{v,c}\Sigma_k 2(P_y^* P_x + P_x^* P_y)\delta$

From our direct calculations of the DT elements we have found that the main mechanism underlying the PA is a sort of ''selection rule'' where only the -110 (for example) polarization directions contribute to the first optical transition while the photons polarized along the 110 direction do not contribute at all. The opposite is true for the second transition. This leads to two different absorption onsets for light polarized along the (110) and (-110) directions, respectively. This is a surprising result, leading potentially to a strong anisotropy effect, in agreement with the experimental data. We have then analyzed the interplay of two factors in determining the PA: the strength of the DT elements on one side and the crystal field splitting of the first two optical transitions, on the other, which are not easily related to each other, as inferred instead in a previous theoretical model.[1]

We start our study by considering the DT which, at the first-order perturbation theory in the dipole approximation is given by:

$$\epsilon_{i,j}(\omega) = \frac{8\pi^2 e^2}{\omega^2 m^2 V} \sum_{v,c} \sum_k (P_{v,c}(k))_i^* (P_{v,c}(k))_j$$
$$\times \delta(E_c(k) - E_v(k) - \hbar\omega), \qquad (2)$$

where $i, j = x, y, z$. $\vec{P}_{v,c}(k) = \langle v, k | \vec{P} | c, k \rangle$ is the matrix element of the optical transition between the valence state $|v, k\rangle$ and the conduction state $|c, k\rangle$. The optical absorption for a wave travelling along the z axis, polarized along a general direction in the xy plane is proportional to

$$\epsilon_2(\omega, \theta) = \epsilon_{xx}(\omega)\cos^2\theta + \epsilon_{xy}(\omega)\sin 2\theta + \epsilon_{yy}(\omega)\sin^2\theta. \qquad (3)$$

We see from this expression that to have in-plane isotropy of the optical absorption the following conditions have to be verified:

$$\epsilon_{xx}(\omega) = \epsilon_{yy}(\omega) \quad \text{and} \quad \epsilon_{xy}(\omega) = 0. \qquad (4)$$

We observe that the dipole oscillator matrix elements transform under the operation of the crystal symmetry group as[5]

$$\vec{P}_{v,c}(R^{-1}\vec{k}) = e^{i\gamma} R \vec{P}_{v,c}(\vec{k}), \qquad (5)$$

where R indicates a symmetry operation of the crystal point group. By applying this property we reduce the summation in Eq. (2) over the only k points in the irreducible wedge of the Brillouin zone (IBZ) and determine the elements of the DT for the CA (D_{2d}) and NCA (C_{2v}) crystal structures. We report the results in Table I. To observe an anisotropy of the optical absorption for a photon polarization along $[1,1,0]$ ($\theta = \pi/4$) and along $[-1,1,0]$ ($\theta = -\pi/4$), $\Delta\epsilon(\omega)$ has to be different from zero, where

$$\Delta\epsilon(\omega) = \epsilon_2\left(\omega, \frac{\pi}{4}\right) - \epsilon_2\left(\omega, -\frac{\pi}{4}\right) = 2(\epsilon_{xy}(\omega)). \qquad (6)$$

$\Delta\epsilon(\omega)$ is clearly zero by symmetry in the CA SL's ($\epsilon_{xy} = 0$, see Table I) but not necessarily in the NCA SL's. We stress here that this fact does not apply only to the particular case of the NCA SL's, but, given the generality of our considerations, it applies to all cases where the point group symmetry reduction leads to $\epsilon_{xy} \neq 0$. A second important consequence of the reduction of the point group symmetry is the CF energy splitting of all the two-degenerate states belonging to the bidimensional E representation of the D_{2d} group into the two unidimensional representations of the C_{2v} group: $E \rightarrow B_1 + B_2$. A consequence of the CF splitting is that there are now two distinct optical transitions instead of one.

To quantify these group theory predictions we have calculated the electronic and optical properties of a $(In_{0.5}Ga_{0.5})As$-InP SL within a first-principles approach. The calculation has been performed in the self-consistent density functional theory, with the local density approximation (LDA), using nonlocal norm-conserving pseudopotentials.[6] The Ceperley Alder form[7] for the exchange and correlation energy has been used. The SRL wave functions are expanded in a plane-wave basis with a 12 Ry energy cutoff. The random alloy $(In_{0.5}Ga_{0.5})As$ has been modeled within the simple virtual crystal approximation. We consider a $[(InGa)As]_n - [InP]_n$ SL with $n = 12$ (corresponding to a quantum well of 35 Å thickness) which is long enough to separate, to an acceptable degree, the contributions coming from the two different interfaces. We study first the simplest possible situation in which we neglect spin-orbit interaction effects and the elastic strain effects at the interfaces. Under these assumptions only the chemical difference of the constituent atoms at the interfaces is taken into account. The calculated DT elements corresponding to the optical transitions between the upper valence states and the first conduction state at the conduction band minimum (CBM) at the Γ point are given in the first part of Table II. We see that only the CF-split $V1$ and $V2$ states contribute to the in-plane absorption since the $V3$-CBM transition has oscillator strength components only along the z axis. From the calculated DT components we see the following: (i) the in-plane elements are fully decoupled from the elements along the z growth direction; (ii) the xy elements of the DT relative to the in-plane transitions are different from zero in the case of the NCA SL's as predicted by theory group symmetry considerations; (iii) very importantly, the xy elements of the DT relative to the CF-split states have opposite signs. From these results we can study the origin of the optical absorption anisotropy with the po-

TABLE II. Calculated dielectric tensor elements for the unstrained $\varepsilon = 0$ and strained $\varepsilon = 0.06$ NCA superlattice.

Transition	V1	V2	V3
		$\varepsilon = 0$	
Energy (eV)	0.6273	0.6288	0.6637
ϵ_{xx}	4.0741	4.0859	0.00
ϵ_{yy}	4.0741	4.0859	0.00
ϵ_{zz}	0.00	0.00	7.1095
ϵ_{xy}	−4.0741	+4.0859	0.00
		$\varepsilon = 0.06$	
Energy (eV)	0.5917	0.6056	0.6781
ϵ_{xx}	4.1016	4.1090	0.00
ϵ_{yy}	4.1016	4.1090	0.00
ϵ_{zz}	0.00	0.00	6.9672
ϵ_{xy}	−4.1016	+4.1090	0.00

larization direction. By considering only the two $V1$ and $V2$ split states at the top of the valence band (VBM) at the Γ point and indicating with ω_1 and ω_2 the transition energies to the CBM state, we see, from the calculated values of the $\epsilon_2(\omega)$ components and from Eq. (3), that

$$\epsilon_2\left(\omega_1, \frac{\pi}{4}\right) = 0, \quad \text{while} \quad \epsilon_2\left(\omega_1, -\frac{\pi}{4}\right) = -2\epsilon_{xy}(\omega_1), \quad (7)$$

$$\epsilon_2\left(\omega_2, \frac{\pi}{4}\right) = 2\epsilon_{xy}(\omega_2), \quad \text{while} \quad \epsilon_2\left(\omega_2, -\frac{\pi}{4}\right) = 0, \quad (8)$$

where $|\epsilon_{xy}| = \epsilon_{xx} = \epsilon_{yy}$. That is, the polarization direction selectively enhances or quenches the optical transitions associated to the C_{2v} CF-split states, i.e., for these transitions taken individually this selection rule gives a *polarization rate of 100%*.[8]

When $\epsilon_{xy}(\omega_1) \approx -\epsilon_{xy}(\omega_2)$ the anisotropy effects tend to subtract in the absorption spectrum, thus the PA is practically *only* due to the small splitting (1.5 meV) $\omega_2 - \omega_1$ between the two CF-split states.[9] The calculated absorption spectrum in the energy range of the two transitions is given in Fig. 1(a) where we see the substantial superposition of the two curves (corresponding to the two polarization directions along $[-110]$ and $[110]$, respectively).[10] The corresponding maximum polarization rate $p(\omega)$, Eq. (1), is small, about 2%. The trend of $p(\omega)$ is decreasing in the energy range between the first and the second transitions, according to the experimental results.

We expect the polarization-dependent anisotropy of the optical absorption to increase by including strain at the interfaces, by increasing the splitting between the two C_{2v}-split states at the VBM, by increasing the off-diagonal elements of the DT relative to the two states, or by both the latter two. We have included strain at the interfaces following the example of Ref. 11. The resulting DT elements are given in the second part of Table II. Strain has reduced the fundamental optical gap and opened further the gap between the two in-plane $V1$ and $V2$ hole states at VBM. The splitting is now 14 meV. We see, however, that the DT elements are almost unchanged because the diminished transition energies are
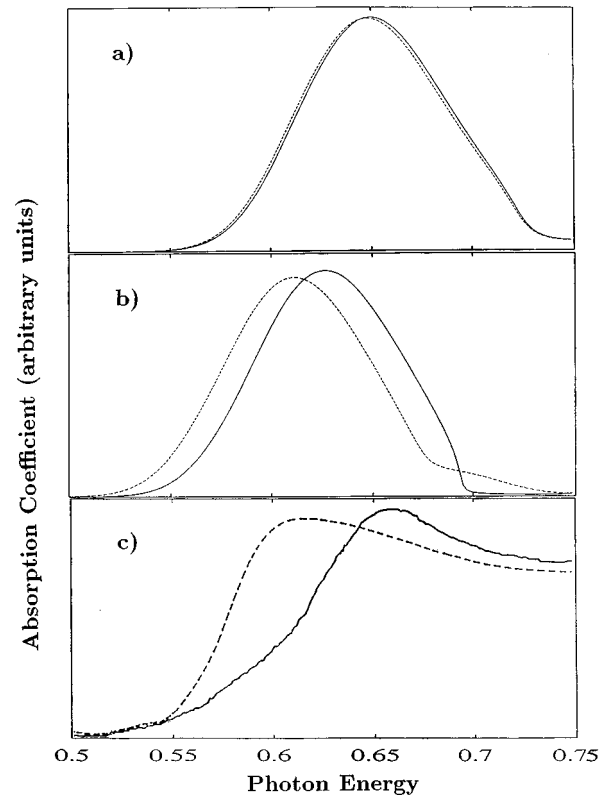

FIG. 1. Calculated absorption spectra for a photon polarization along $[-110]$ (dashed curve) and along $[110]$ (continuous curve) for unstrained interfaces (a) and strained interfaces (b), compared to the experimental results taken from Ref. 3, (c). In (c) the x axis has been rigidly shifted by 325 meV towards lower energies to have the experimental curves over the same energy range of the calculated ones. Of the 13 k points included in the calculation (see Ref. 10) only the Γ point has transitions falling in the considered energy range. To reproduce the tails of the experimental features it would have been necessary to include many more k points near Γ in the calculation.

compensated by reduced dipole oscillator strengths. Again, absorption corresponding to a polarization vector along the $[110]$ direction selects one transition, while absorption corresponding to a polarization vector along $[-110]$ ''sees'' only the other. The mechanism of the anisotropy is therefore disclosed entirely by the further splitting of the two hole states. The absorption peaks corresponding to the two transitions are now fully resolved. The spectrum is given in Fig. 1(b). Inclusion of the strain at the interfaces has increased the maximum polarization rate $p(\omega)$, now 15%, by an order of magnitude. Comparing the spectrum of Fig. 1(b) with the experimental absorption spectra in Fig. 1(c), we find a striking resemblance. The similarity, both in experiment and theory, in the shape and intensity of the two absorption spectra for polarizations along $[110]$ and $[-110]$, respectively, reflects the similarity of the two optical transitions oscillator strengths; moreover, we find that the peak associated with the second transition (higher in energy) is slightly more pronounced than the first one, in agreement with the experiment. The shift between the two absorption features displayed by our calculated spectra turns out to be still underestimated (14 meV against the experimental value, about 40 meV), thus

$p(\omega)$ is still underestimated. It is interesting to note that, unlike the model of Ref. 1, the polarization rate here is directly linked to the energy gap between the splitted states: a gap of 1.5 meV gives a $p(\omega)$ maximum of $\sim 2\%$, whereas a gap of 14 meV increases this value to $\sim 15\%$. Obviously we have still neglected other important effects which could contribute to split further the two hole states at VBM, such as spin-orbit coupling, a more complete atomic relaxation of the entire superlattice, and, possibly, a quasiparticle correction to the LDA hole eigenenergies. The physical description of PA in our work is different from that of Refs. 1 and 3, as the full, nonperturbative, treatment of the SRL potential leads to a different relation between the strength of the DT elements and the splitting between the two upper hole states at VBM, and leads also to the giant effect related to the ''selection rule'' between the components of the doublet. These effects are experimentally resolved at the onset of the absorption.

In conclusion we have shown how symmetry reduction in NCA SL's leads to a polarization-dependent anisotropy of the optical absorption. We have found that the polarization direction selects (that is, enhances or quenches) the optical transitions to the CBM from the valence eigenstates which are degenerate in CA SL's. This mechanisms gives rise to the observed anisotropy with photon polarization and is likely to be present also in other systems with broken degeneracies. Our results suggest ways to enhance the anisotropy through manipulation of the interface structure or chemistry or strain condition.

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[8] It should be noted that, due to our z-axis orientation with respect to the bonds orientation at the interfaces, the 110 direction in our calculation corresponds to the -110 in the experiment of Ref. 3 and vice versa.

[9] The results $|\epsilon_{xy}| = \epsilon_{xx} = \epsilon_{yy}$ and $\epsilon_{xy}(\omega_1) \approx \epsilon_{xy}(\omega_2)$ are true not only at Γ for the split states. They are practically verified for every k point in the vicinity of the zone center, playing a role for the absorption in the energy range of the observed anisotropy.

[10] Thirteen k points in the IBZ have been included in the calculation of the absorption. The calculated $\alpha(\omega)$ has been dressed by a Gaussian broadening of 30 meV. The ~ 0.3 eV discrepancy between theoretical and experimental absorption onset is due to the use of LDA.

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