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# Observation of strong anisotropic forbidden transitions in (001) InGaAs/GaAs single-quantum well by reflectance-difference spectroscopy and its behavior under uniaxial strain

Jin-Ling Yu, Yong-Hai Chen\*, Chen-Guang Tang, ChongYun Jiang, Xiao-Ling Ye

### **Abstract**

The strong anisotropic forbidden transition has been observed in a series of InGaAs/GaAs single-quantum well with well width ranging between 3 nm and 7 nm at 80 K. Numerical calculations within the envelope function framework have been performed to analyze the origin of the optical anisotropic forbidden transition. It is found that the optical anisotropy of this transition can be mainly attributed to indium segregation effect. The effect of uniaxial strain on in-plane optical anisotropy (IPOA) is also investigated. The IPOA of the forbidden transition changes little with strain, while that of the allowed transition shows a linear dependence on strain.

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### Introduction

It is well known that in-plane optical anisotropy (IPOA) can be introduced in a (001)-grown zinc-blende quantum well (QW) when the symmetry is reduced from  $D_{2d}$  to  $C_{2\upsilon}$  [1-6]. There are two kinds of symmetry reduction effect (SRE), one is bulk SRE, and the other is interface SRE [2,4]. The bulk SRE can be introduced by electric field, compositional variation across the QW and uniaxial strain [7-10]. The IPOA induced by uniaxial strain in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QWs has been reported by Shen [10], Rau [8] and Tang [11]. However, as far as we know, this effect in  $In_xGa_{1-x}As/GaAs$  QW has never been reported.

The interface SRE, which origins from  $C_{2v}$  symmetry of a (001) zinc-blende interface, can be introduced by special interface chemical bonds, segregation effect and the anisotropic interface structures [2,3,6]. It was found that the interface-induced IPOA was very strong in the QWs sharing no-common-atom, while the IPOA in QWs sharing common atoms such as GaAs/AlGaAs was too weak to be observed by conventional polarized spectroscopy [2,4,10]. Fortunately, the weak IPOA in the AlGaAs/

GaAs and InGaAs/GaAs QWs can be well observed by the reflectance-difference spectroscopy (RDS) [2,4,6]. Wang et al. has studied forbidden transitions in In<sub>x</sub>Ga<sub>1-</sub> <sub>x</sub>As/GaAs by photoreflectance (PR) and attributed the forbidden transition to the built-in electric field [12]. Chen et al. [1] and Ye et al. [6] observed anisotropic forbidden transition in In<sub>x</sub>Ga<sub>1-x</sub>As/GaAs by RDS. Chen ascribed the anisotropic forbidden transition to the interplay of interface  $C_{2\nu}$  symmetry and built-in electric field, while Ye attributed it to both the built-in electric field and segregation effect. In this study, we observed strong anisotropic forbidden transitions in a series of In<sub>x</sub>Ga<sub>1</sub>-<sub>x</sub>As/GaAs single-quantum well (SQW) with well width ranging between 3 nm and 7 nm at 80 K. Numerical calculation within the envelope function framework have been performed to analyze the origin of the optical anisotropic forbidden transition. Detailed theory-experiment comparisons show that the anisotropic forbidden transition can be mainly attributed to indium (In) segregation effect. Besides, the effect of uniaxial strain on in-plane optical anisotropy (IPOA) is also investigated. It is found that, the IPOA of the forbidden transition nearly does not change with strain, while that of the allowed transition shows a linear dependence on strain. Finally, an

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interpretation of the IPOA by perturbation theory is given out.

### Samples and experiments

A series of In<sub>0.2</sub>Ga<sub>0.8</sub>As/GaAs SQW with different well widths were grown on (001) semi-insulating GaAs by molecular beam epitaxy. The SQW was sandwiched between two thick GaAs layers. The nominal well widths of the three samples were 3, 5, and 7 nm, respectively. All epilayers were intentionally undoped. The setup of our RDS, described in Ref. [13], is almost the same as Aspnes et al. [14], except the position of the monochromator. The relative reflectance difference between [110] and [110] directions, defined by  $\Delta r/r = 2$  $(r_{110} - r_{110})/(r_{[110]} + r_{[110]})$ , was measured by RDS at 80 K. Here  $r_{[110]}$  ( $r_{[1\underline{1}0]}$ ) is the reflective index in the [110] ([110]) direction. We also did the reflectance measurements, and thus obtained the  $\Delta R/R$  spectra. Here R is the reflectivity of the sample and  $\Delta R$  is the reflectivity difference between samples with and without QW layer.

In order to study the effect of uniaxial strain on the IPOA, we cleaved the sample with well width 5 nm into a  $25 \times 4$  mm<sup>2</sup> strip. Uniaxial strain was introduced by a stress device as shown in Figure 1 which is the same as the one used by Papadimitriou and Richter [15]. When the length-to-width ratio is greater than 3, the strip behaves like a bend rod, and the apparatus produces only two nonzero strain components: $\epsilon_{x'x'}$  (tensile)  $\epsilon_{z'z'}$  and (compressive). Here x' and y' are along the cleavage axis [110] and [110] as shown in Figure 1. Transformed to the principal axis [100] and [010], the nonzero strain components are  $\epsilon_{xx}$ ,  $\epsilon_{yy}$ ,  $\epsilon_{zz}$  and  $\epsilon_{xy}$  [4], and only  $\epsilon_{xy}$  will

introduce IPOA. The maximum strain component  $\epsilon_{xy}$  at the center of the strip is given by [16]

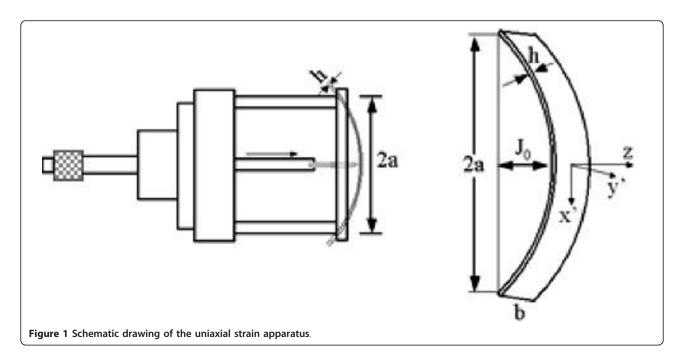
$$\varepsilon_{xy} = \frac{\varepsilon_{x'x'}}{2} = \frac{3hJ_0}{4a^2}.$$

Here  $J_0$  is the deformation at the strip center, h is the thickness and 2a is the length of the strip. The relative reflectance difference between the [110] and [110] directions at the center of the strip (3 × 4 mm<sup>2</sup>) is measured by RDS at room temperature.

# Results and discussion

### **Experimental results**

Figure 2 shows the real part of the RD and  $\Delta R/R$  spectra of the three samples obtained at 80 K. In the  $\Delta R/R$  spectra, we can observe the transitions of 1e1hh (the first conduction to the first valence subband of heavy hole), 1e1lh and 2e2hh, and what's more, the intensity of the transition 1e1hh is much larger than that of the 1e1lh. However, in the RD spectra, besides the allowed transitions 1e1hh, 1e1lh, 2e2hh and 1eh\*, we can also observe the forbidden transition 1e2hh. Here h\* represents continuous hole states. The energy positions of the transitions 1e1hh (1e1lh) are marked by solid (dotted) lines. And the positions of 1e2hh, 1eh\* and 2e2hh are indicated by upward, green downward and black downward arrows, respectively. The transitions 1e1hh and 1e1lh show peak-like lineshape (negative or positive), while the forbidden transitions 1e2hh of the samples with well width 5 and 7 nm present a smoothed-step-like lineshape. This phenomenon may be attributed to the



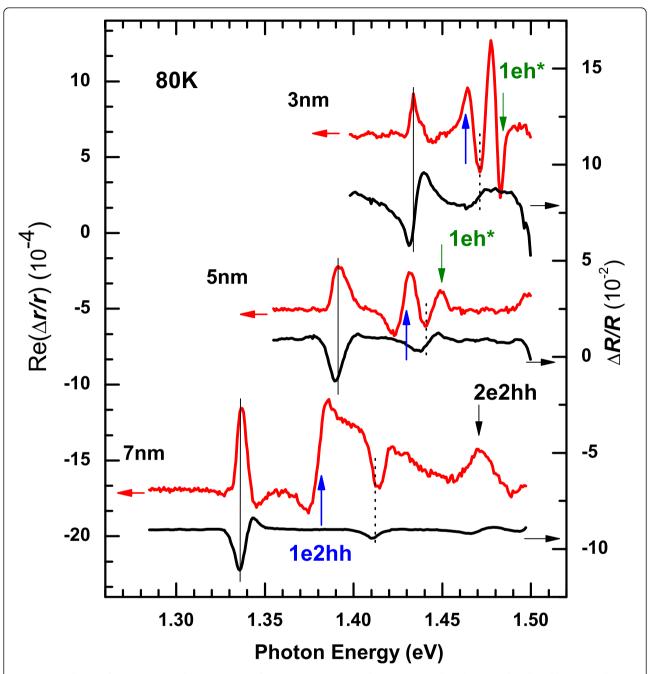


Figure 2 Real part of RD spectra and  $\Delta R/R$  spectra of  $In_{0.2}Ga_{0.8}As/GaAs$  single-quantum well with nominal well width 3, 5, and 7 nm, respectively. The spectra are measured at 80 K. The vertical lines indicate the energy positions of the transitions 1e1hh (solid) and 1e1lh (dotted). And the vertical arrows indicate the positions of 1e2hh (upward arrows), leh\* (downward arrows), and 2e2hh (downward arrows). Here h\* represents continuous hole states.

coupling of heavy and light holes when the in-plane wave vector is nonzero [1]. For the sample with well width 3 nm, it is difficult to clearly distinguish the corresponding energy positions of the transitions 1e2hh, 1e1lh and leh\*, because they are too close to each other. Even so, we can still observe that, the intensity of the IPOA of 1e1lh increases obviously compared to that of

1e1hh. Surprisingly, the forbidden transition 1e2hh are comparable to the allowed transition in RD spectra, while it almost cannot be observed in  $\Delta R/R$  spectra.

Figure 3 shows the imaginary part of RD spectra of the sample with 5 nm well width under different strain. Although the signal-to-noise ratio at room temperate is not as good as that at 80 K, three structures can still be

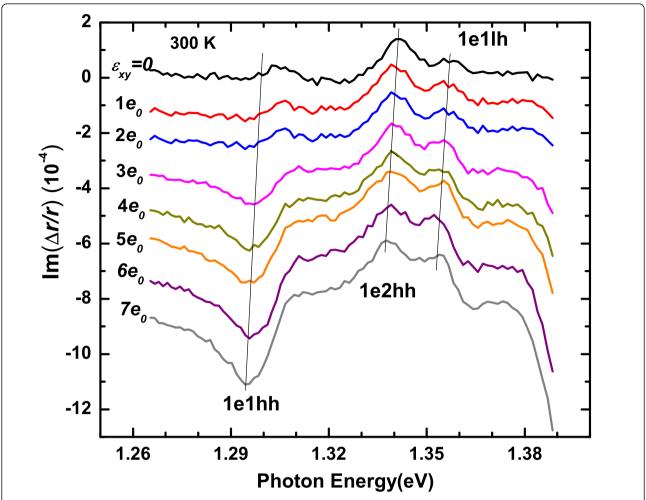


Figure 3 RD spectra of 5 nm-ln<sub>0.2</sub>Ga<sub>0.8</sub>As/GaAs QW under different strain  $\epsilon_{xy}$  in unit of  $\epsilon_0 = 3.23 \times 10^{-5}$ . The spectra are measured at room temperature and shifted vertically for clarity. The oblique lines indicate the energy positions of the transitions 1e1hh, 1e2hh, and 1e1lh in the RD spectra.

clearly observed in the vicinity of 1.30, 1.34 and 1.36 eV, which can be assigned to the transitions of 1e1hh, 1e2hh and 1e1lh, respectively. Figure 4a shows us the RD intensity of the transition 1e1hh, 1e2hh and 1e1lh vs. strain, after subtracting the RD contribution under zero strain. It can be seen that, as the strain increases, the RD intensity of the allowed transition 1e1hh and 1e1lh are enhanced, while that of the forbidden transition 1e2hh does not show apparent change. Besides, in contrast to the transition 1e2hh and 1e1lh, the sign of the anisotropic transition 1e1hh changes as the strain increases. In addiction, slight redshifts can be introduced by the strain for all transitions, as shown in Figure 4b. The energy shift caused by  $J_0 = 0.07$  (i.e.,  $\epsilon_{xy} = 7e_0 = 2.3 \times 10^{-4}$ ) is less than 9 meV.

### Models and calculation results

It is well known that, IPOA in (001)-oriented QWs mainly comes from mixing between heavy and light

holes [2,3,17]. However, it is demonstrated that the spin-orbit coupling has significant effects on the band structure especially for highly strained quantum wells [18]. The strain will couple the heavy-hole (hh) bands, light-hole (lh) bands with spin-orbit split-off (SO) band [18]. Therefore, taking into account the coupling between hh, lh and SO band, we use 6 band  $\mathbf{K} \cdot \mathbf{P}$  theory which is described in Ref. [18], and treat the holemixing induced by the strain  $\epsilon_{xy}$ , electric field and the two interface as perturbation [4]. The perturbation Hamiltonian H can be written as [18]

$$H' = \begin{pmatrix} 0 & 0 & iR & 0 & 0 & iR \\ 0 & 0 & 0 & iR & Q & 0 \\ -iR^{\dagger} & 0 & 0 & 0 & 0 & Q \\ 0 & -iR^{\dagger} & 0 & 0 -iR^{\dagger} & 0 \\ 0 & Q & 0 & iR & 0 & 0 \\ -iR^{\dagger} & 0 & Q & 0 & 0 & 0 \end{pmatrix}$$
 (1)

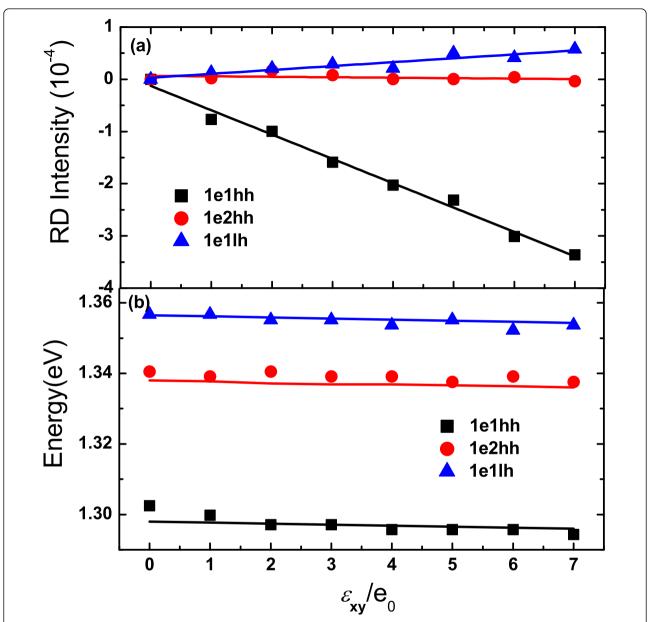


Figure 4 Strain dependence of RD intensity and energies of 1e1hh, 1e2hh and 1e1lh. (a) RD intensity of the transitions 1e1hh (squares), 1e2hh (circles) and 1e1lh (triangles) vs. strain after subtracting the RD contribution under zero strain. The solid lines are the linear fitting of the experimental data. (b) The transition energies vs. strain. The solid lines in (b) are calculated from the envelope function theory  $(1e_0 = 3.23 \times 10^{-5})$ 

with [2,4]

$$R(z) = \left\{ Dd_{14}F + D\varepsilon_{xy} + \left[ \frac{P_1}{l_1} \exp(-\frac{z - w/2}{l_1})\Theta(z - w/2) - \frac{P_2}{l_2} \exp(-\frac{z + w/2}{l_2})\Theta(z + w/2) \right] \right\},$$
(2)

and [18]

$$Q = -\frac{b}{2} (\varepsilon_{xx} + \varepsilon_{\gamma\gamma} - 2\varepsilon_{zz})$$
 (3)

for the basis  $|3/2, 3/2\rangle$ ,  $|3/2, 1/2\rangle$ ,  $|3/2, -1/2\rangle$ ,  $|3/2, -3/2\rangle$ ,  $|1/2, 1/2\rangle$ , and  $|1/2, -1/2\rangle$ . Here b and D are the Bir-Pikus deformation potentials, F is the electric field along the z direction,  $d_{14}$  is the piezoelectric constant,  $\epsilon_{ij}$  denotes the symmetric strain tensor,  $P_1$  ( $P_2$ ) is the lower (upper) interface potential parameter describing the effect of  $C_{2v}$  interface symmetry [2],  $l_1$  ( $l_2$ ) is the In segregation length in the lower (upper) interface, and  $z=\pm w/2$  is the location of the interfaces of QW. The interface potential parameter  $P_1$  and  $P_2$  are equal for a

symmetric QW, and anisotropic interface roughness will make them unequal [4]. According to the model suggested in Ref. [19], we assume that the segregation lengths on the two interfaces are equal, i.e.,  $l_1 = l_2$ .

In order to estimate the value of built-in electric field, we perform photoreflectance measurements. However, no Franz-Keldysh oscillations presents, which can be attributed to the fact that the layers are all intentionally undoped and the residual doping is very low. Thus, the residual electric field is weak enough to be neglected.

Based on the Luttinger  $6 \times 6$  hole Hamiltonian [18] and the hole-mixing Hamiltonian described in Equation 1, the energies of ne-mlh/hh transition and transition probability can be calculated. Then using a Lorentzian function, as described in Equation 4, we can simulate anisotropic transition spectroscopy  $\Delta M$  and average transition spectroscopy M.

$$M(or \, \Delta M) = \sum_{n,m} \frac{1}{\pi} \frac{0.5 \, \Gamma}{\left(E - E_{nm}\right)^2 + \left(0.5 \, \Gamma\right)^2} \times P_{nm}, \quad (4)$$

here  $\Gamma$  is the linewidth of the transition, and  $E_{nm}$  ( $P_{nm}$ ) is the transition energy (probability) between ne and mlh or between ne and mhh. In the calculation, the adopted Luttinger parameters are:  $\gamma_1 = 6.85$ ,  $\gamma_2 = 1.9$ ,  $\gamma_3$ = 2.93 for GaAs, and  $\gamma_1$  = 21.0,  $\gamma_2$  = 8.3,  $\gamma_3$  = 9.2 for InAs. The band-offset is taken as Qc = 0.64 [20], and the strain-free In<sub>x</sub>Ga<sub>1-x</sub>As band gap at 80 and 300 K are taken from Refs. [20] and [21], respectively. The other band parameters are got from Ref. [22]. The anisotropic transition probability  $\Delta M$  is proportional to  $\Delta r/r$ . Therefore, we can compare the theoretical calculated  $\Delta M$ with experimental data  $\Delta r/r$ , and thus to find out the reason responsible for the observed strong anisotropic forbidden transitions. It is noteworthy that even under zero uniaxial strain, there will still be residual anisotropic strain exists, which may be due to a preferred distribution of In atoms [23]. In the following, we will discuss the interface potential, segregation and anisotropic strain effect separately.

We should first estimate the value of interface potential parameter, denoted as  $P_0$ . So far, there are four theoretical models estimating the value of  $P_0$ : boundary conditions (BC) model by Ivchenko [17], perturbed interface potential model (called " $H_{BF}$ ") by Krebs [3], averaged hybrid energy (AHE) difference of interfaces model and lattice mismatch model by Chen [24]. Given that BC model is equivalent to  $H_{BF}$  model, we need to consider only one of them [24]. Thus using  $H_{BF}$ , AHE and lattice mismatch model and then adding them up, we obtain the value of  $P_0$  is about 600 meV Å.

If there is only anisotropic interface structures in the interface, i.e., l=0,  $\epsilon_{xy}=0$ , we can adopt  $P_1=P_0$ , and fit  $P_2$  to the experimental data. The fitting results are

shown in Figure 5a. The  $P_2$  value adopted is 775 meV Å. It can be seen that, only the allowed transition presents. Therefore, the observed anisotropic forbidden transition cannot be attributed to anisotropic interface structures.

If there is only anisotropic strain effect in the QW (i.e.,  $P_1 = P_2 = P_0$ , l = 0), only one free parameter  $\epsilon_{xy}$  can be fitted to the experimental data. The fitting result is shown in Figure 5b. The  $\epsilon_{xy}$  value we adopt is  $0.003 \times \epsilon_{xx} = -4.24 \times 10^{-5}$ . Again, there is no forbidden transition presents. Therefore, the observed anisotropic forbidden transition cannot be attributed to anisotropic strain effect.

If there is only atomic segregation effect (i.e.,  $P_1 = P_2 = P_0$ ,  $\epsilon_{xy} = 0$ ), one can fit free parameter l to the experimental data. The fitting result is shown in Figure 5c. The fitted segregation length l is 1.8 nm, which is in reasonable agreement with that reported in Ref. [19]. Apparently, the segregation effect will lead to a strong IPOA for the forbidden transition 1e2hh, but do not change its average transition probability, which is still very small. Besides, for the sample with well width 3 nm, a strong IPOA is also present for the transition 1e1lh. Therefore, the observed anisotropic forbidden transition is closely related to In atomic segregation effect.

From Figure 5c, we can see that, if there is only segregation effect, the sign of the transition 1e1hh is negative, which is not consistent with the experiment. Therefore, there must be some other effect existing, such as anisotropic interface structures or anisotropic strain effect. When we take both the anisotropic strain and segregation effect into account, the calculated results are not consistent with the experimental data. However, the results obtained by both the anisotropic interface structure and the segregation effect are in reasonable agreement with the experiment, as shown in Figure 5d. In the calculation, we adopt interface parameter  $P_1 = 595$ meV Å,  $P_2$  = 775 meV Å, and the segregation length l = 1.8 nm. The obtained interface potential difference  $\Delta P/$  $P_0$  is about 30%, which is much larger than that obtained in GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As QW (about 6%) [4]. The reason may be that lattice mismatch will enhance the interface asymmetry of the QWs.

Using the parameters obtained above, we can well stimulate the IPOA of all the transitions under different uniaxial strain, as shown in Figure 6. The calculated transition energies are also well consistent with experiments, which is shown in Figure 4b.

### Interpretation of IPOA by perturbation theory

The IPOA-intensity ratio of 1e1lh and 1e1hh transitions is much stronger for the sample with 3 nm well width compared to that of the other samples. This phenomenon may

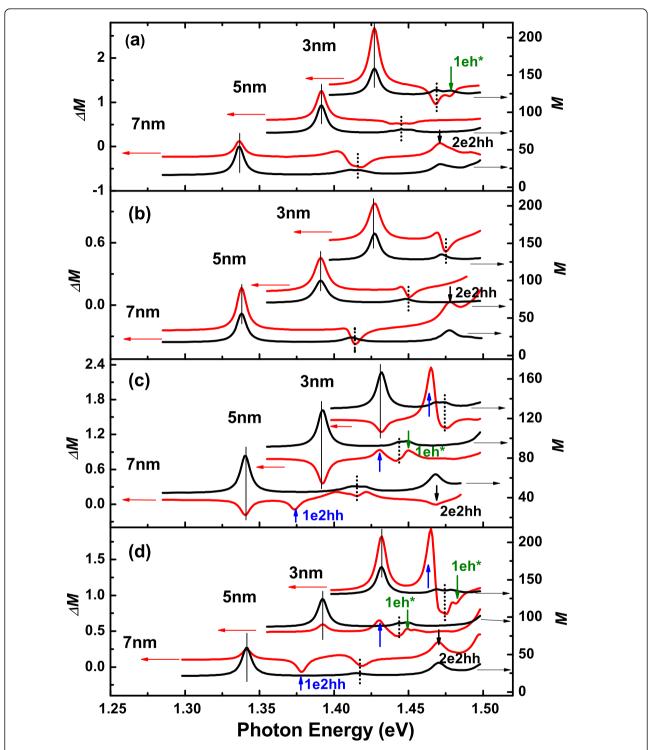


Figure 5 Calculated anisotropic transition probability  $\Delta M$  and average transition probability M of  $\ln_x Ga_{1-x} As/GaAs$  QW with well width 3, 5 and 7 nm, respectively. The optical anisotropy is induced by (a) anisotropic interface structures, (b) anisotropic strain effect, (c) In segregation effect and (d) both anisotropic interface structures and In segregation effect. The vertical lines indicate the energy positions of the transitions 1e1hh (solid) and 1e1lh (dotted). And the vertical arrows indicate the positions of transitions 1e2hh (upward arrows), leh\* (downward arrows), and 2e2hh (downward arrows).

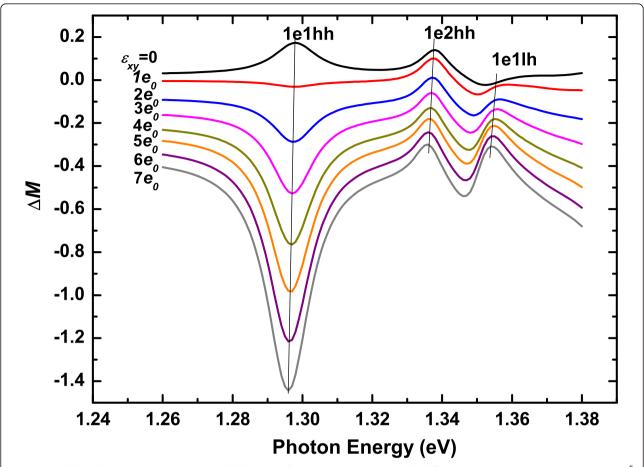


Figure 6 Calculated anisotropic transition probability  $\Delta M$  of  $\ln_x \text{Ga}_{1-x} \text{As/GaAs}$  QW under different strain  $\epsilon_{xy}$  in unit of  $e_0 = 3.23 \times 10^{-5}$ . The oblique lines indicate the energy positions of the transitions 1e1hh, 1e2hh, and 1e1lh in the  $\Delta M$  spectra.

be undefirstood in the following way. According to perturbation theory, the anisotropic transition probability  $\Delta M$  of 1e1lh can be expressed as [1,2]

$$\frac{\langle 1E|1H\rangle\langle 1H|R(z)|1L\rangle\langle 1L|1E\rangle}{E_{1L}-E_{1H}} + \frac{\langle 1E|2H\rangle\langle 2H|R(z)|1L\rangle\langle 1L|1E\rangle}{E_{1L}-E_{2H}}.$$
(5)

Here  $\langle 1E|nH\rangle$  is the overlap integral between the first electron and the nth heavy-hole states.  $\langle 1H|R(z)|1L\rangle$  is the hole-mixing strength between 1H and 1L.  $E_{1L}$  -  $E_{nH}$  is the energy separation between 1L and nH. It can bee seen that,  $\Delta M$  is directly proportional to the coupling strength of holes and inversely proportional to their energy separation. For the three samples, there is little difference in the term R(z). However,  $E_{1L}$  -  $E_{2H}$  of the sample with 3 nm well width is smaller than that of the other samples, which results in much stronger IPOA.

The appearance of the forbidden transition and its behavior under uniaxial strain can be interpreted in a similar way. According to perturbation theory, the anisotropic transition probability  $\Delta M$  of 1e2hh can be expressed as [1,2]

$$\frac{\langle 1E|1L\rangle\langle 1L|R(z)|2H\rangle\langle 2H|1E\rangle}{E_{2H} - E_{1L}} + \frac{\langle 1E|2H\rangle\langle 2H|R(z)|SO\rangle\langle SO|1E\rangle}{E_{2H} - E_{SO}}.$$
(6)

Here  $\langle 1E|nH\rangle$  and  $\langle nH|1E\rangle$  ( $\langle {\rm SO}|1E\rangle$ ) are the overlap integrals between the discussed electron and hole (SO) states.  $\langle 1L|R(z)|2H\rangle$  is the hole-mixing strength between 1L and 2H, and  $\langle 2H|R(z)|{\rm SO}\rangle$  is the coupling strength between 2H and SO band.  $E_{2H}$  -  $E_{{\rm SO}}\gg E_{{\rm 2H}}$  -  $E_{{\rm 1L}}$ , the coupling between 1L and 2H dominates. When there is no segregation effect,  $\langle 2H|1E\rangle=0$  and no optical anisotropy exists. However, when segregation emerges, the symmetric square well changes into an asymmetric well, which will change the parities of the subband wave functions. Besides, it will also couples the 1L and the 2H subbands, and as a result, the perturbed 2H subband wave function now contains a small portion

of the unperturbed  $|1L\rangle$  one. Thus,  $\langle 2H|1E\rangle \neq 0$ , and its value is proportional to the segregation effect. The strain component  $\epsilon_{xy}$ , being an even function of space, only couples the sub-bands with same parity, such as 1H and 1L. Then, the contribution of  $\epsilon_{xy}$  to the numerator of the first term in Equation 6 can be written as

$$\langle 1E|1L\rangle D\varepsilon_{xy}\langle 1L|2H\rangle\langle 2H|1E\rangle,$$
 (7)

in which the first integral is nearly a constant, and  $\langle 1L|2H\rangle$   $\langle 2H|1E\rangle$  is mainly determined by the segregation effect and interface potential. Therefore, for the forbidden transition 1e2hh, the change of IPOA induced by a weak uniaxial strain (in the order of  $10^{-5}$ ) will be too weak to be observed in experiment. However, for the allowed transitions, such as 1e1hh, the strain will also couple 1H and 1L, and will remarkably change the IPOA. From Figure 3 we can see that the RD intensity of transition 1e1lh does not show significant change as the strain increases. The reason may be that the lighthole band configuration is weak type I for the current alloy composition [20], which result in the change of the potential has little influence on its wave function.

### Conclusion

We have observed strong anisotropic forbidden transition in a series of  ${\rm In_{0\cdot 2}Ga_{0\cdot 8}As/GaAs}$  SQW with well width ranging between 3 nm and 7 nm at 80 K. Using a 6 band  $K\cdot P$  theory, we have calculated the optical anisotropy induced by interface composition profile due to In segregation, anisotropic interface structures and anisotropic strain. It is found that the observed anisotropic forbidden transition can be mainly attributed to the In segregation effect. Besides, the effect of uniaxial strain on IPOA is also investigated. It is found that the IPOA of the forbidden transition changes little with strain, while that of the allowed transition shows a linear dependence on strain. Finally, an interpretation of IPOA by perturbation theory is also given out.

### **Abbreviations**

AHE: averaged hybrid energy; BC: boundary conditions; In: indium; IPOA: inplane optical anisotropy; SQW: single-quantum well; SRE: symmetry reduction effect; PR: photoreflectance; QW: quantum well; RDS: reflectancedifference spectroscopy.

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### Authors' contributions

JLY performed the statistical analysis, carried out the calculations and drafted the manuscript. YHC conceived of the study, and participated in its design and coordination. CGT carried out the experiments. CYJ participated in the revision of the manuscript and discussed analysis. XLY participated in the design of the study. All authors read and approved the final manuscript.

### Competing interests

The authors declare that they have no competing interests.

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