

1,2 One of them, discussed here, is the intermediate-band solar cell

(c) the nonradiative recombination of IB electrons with VB holes (process 2); and

(d) the ever-present recombination of CB electrons with VB holes (process 3) must be as slow as possible.

It has been suggested that the IB be made either by alloying a sublattice into a bulk crystal using an early  $3d$  transition metal (TM), e.g., Ti in GaP,<sup>4</sup> Cr in ZnS,<sup>5</sup> and TM in CuGaS<sub>2</sub>,<sup>6</sup> In<sub>2</sub>S<sub>3</sub>, and MgIn<sub>2</sub>SofD

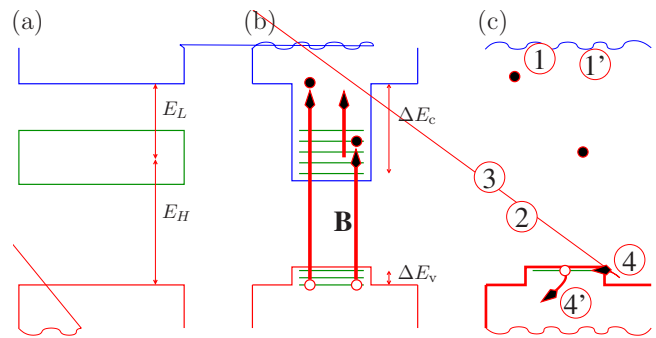


FIG. 1. (Color online) Schematic diagram of single-particle energy levels in a QD array and its equivalent intermediate-band system. (a) Valence band (VB), conduction band (CB), and intermediate-band (IB) energy levels. (b) Schematic of a quantum dot (QD) array with energy levels and transitions labeled 1, 2, 3, 4, 4', and B. (c) Schematic of the equivalent intermediate-band system with energy levels 1, 1', 2, 3, 4, and 4'.

' ) or of dot holes to the matrix VB (process 4');

*al.*<sup>12</sup> (GaSb QDs in GaAs), all these using *p*-GaAs as substrate. It is obvious from condition (i) above that to ensure strong VB  $\rightarrow$  IB and IB  $\rightarrow$  CB absorptions, one needs a high concentration of defect-free dots in the matrix. In the Stranski-Krastanov growth, this implies a significant number of layers of dots separated from each other by the barrier material. However, in the applications mentioned above, the number of dot layers was severely limited by the strain accumulated in the system as the number of dot layers is increased. It was recently proposed<sup>13,14</sup> to introduce GaP strain-compensating layers into the GaAs matrix. A higher efficiency of these cells was obtained, but still the number of QD layers was limited. Based on a similar principle, we explore here a strain-symmetrized architecture, allowing the growth of a large number of dot layers.

Having selected a system that ensures strain symmetrization [a step toward condition (i)], we next examine condition (ii) by measuring and calculating the spectra. We find that the intensity of absorption between QD-confined electron states and the host CB is weak because of their localized-to-delocalized character. Regarding condition (iii) we conclude that, whereas strain symmetrization can indeed allow the growth of multiple dot layers, the current reperto

4.e-613.9(f)TJT\*TD(t)/F4eo)6TD(t)/F4eo)6TD(t)613bya8CB)5ksw/F21Tf0.33

$$\sigma = \frac{U}{\varepsilon} = \frac{1}{V} \frac{E}{\varepsilon}. \quad (1)$$

$\text{In}_{0.47}\text{Ga}_{0.53}\text{As}/\text{GaAs}_{1-x}\text{P}$

We compare in Table II the AE strain-balance predictions for the two pure binaries QW epitaxial systems  $(\text{GaP})_{n_1}/(\text{InAs})_{n_2}$  on GaAs(111) and  $(\text{InAs})_{n_1}/(\text{GaAs})_{n_2}$  on InP(111) compared with the CE estimation, Eq. (6). We used the same VFF force constants and elastic coefficients as for the (001) direction, listed in Table I. As in the case of the (001)-oriented substrate, the AE results depend on the thickness  $n_1 a_1$  of the layer kept fixed. One can also see that the estimations of strain-balanced  $n_2 a_2/n_1 a_1$  ratio obtained using the CE-derived condition are about 2–9 % off the direct AE calculations.

For some orientations, such as (110), the stress tensor is not isotropic, i.e.,  $\sigma$

regular columns of QDs aligned a few degrees off the [113]B growth direction.<sup>31</sup>

The P content in the  $\text{GaAs}_{1-x}\text{P}_x$  barrier layers was varied from  $\sim 0.8\%$  to  $\sim 18\%$  by increasing the phosphine flow during growth. The phosphine flow required for achieving the required  $\text{GaAs}_{1-x}\text{P}_x$  barrier layer compositions was determined by first growing a series of calibration layers on (001) GaAs substrates whose P concentrations were then deduced from x-ray-diffraction measurements.

Our AE strain-balance calculations predict for the  $\text{GaAs}_{1-x}\text{P}_x$  barrier with  $x=0.14$  a thickness  $t_S$  of 32–33 ML. The right panel of Fig. 2

cell containing 14% P, 0.84 V, resulting in a reduced cell efficiency. This may reflect that the cell containing 18% P is no longer strain balanced and contains a higher density of

higher energy,  $\approx 1.51$  eV, reflecting the increase in band gap of the  $\text{GaAs}_{1-x}\text{P}_x$



tice of very high quality can be grown. The beneficial role of balancing the strain in the structure was also seen in the  $I$ - $V$  characteristic and photoluminescence response of the samples, comparing the results at and off the strain-balance matrix composition.

We will investigate in the following the electronic struc-

apply the configuration-interaction (CI) method described in Ref. 40. The excitonic levels can be obtained by diagonalizing the matrix

$$\mathcal{H}_{eh,e'h'} = (E^e - E^h) \delta_{e,e'} \delta_{h,h'} - J_{eh,e'h'} + K_{eh,e'h'}, \quad (19)$$

where  $J$  and  $K$  are the Coulomb and exchange integrals:<sup>40</sup>

$$J_{eh,e'h'} = e^2 \sum_{\sigma_1}$$

and Bloch character of the dot-confined states in the  $\text{In}_{0.47}\text{Ga}_{0.53}\text{As}/\text{GaAs}_{0.86}\text{P}_{0.14}$  system is the same as determined in  $\text{In}_{0.60}\text{Ga}_{0.40}\text{As}/\text{GaAs}$  QDs of comparable height.<sup>41</sup> For the former system, however, the hole states exhibit a smaller heavy hole–light hole mixing.

## *2. Energy separation of the IB from the band edges*

We also show in Fig. 10 explicit values for the energy separations of importance for the QD-IBSC: the energy offsets of the first electron and hole dot-confined levels,  $\Delta E_c = E_c - E_0^e$ , and  $\Delta E_v = E_0^h - E_v$ , as well as the position of  $e_0$  (the intermediate band) within the matrix band gap, corresponding to condition (iii)

benchmark QD-IBSC is shown in Fig. 13(a). We see in this spectrum three major peaks: (i) the first one, at  $\approx 1.32$  eV, corresponds to the intradot  $S_h \rightarrow S_e$  transition; (ii) the second one, at  $\approx 1.40$ – $1.44$  eV, involves dot-confined hole states located still above the matrix VBM, having the proper symmetry allowing transitions to  $S_e$ ; (iii) a broad feature at  $\approx 1.52$ – $1.56$  eV which corresponds to hole states localized

Fig. 14. One notes that variations in P composition have only a small effect (several meV) on the *absolute* position of the dot levels (electron and holes). Because of the strong composition dependence of the matrix CBM, however, the number of confined electron levels increases with increasing  $x$ . Their offset  $\Delta E_c = E_c - E_0^e$  with respect to the matrix CBM also increases with  $x$ , from 48 meV for  $x=0.008$  up to 96 meV for

It is important to mention that this trend includes two effects: (i) the change in VBM and CBM of  $\text{GaAs}_{1-x}\text{P}_x$  with  $x$  in the absence of strain (black lines in Fig. 15) and (ii) the combined chemical and strain effect [red (gray) line]. The chemical effect alone moves  $E_v$  down and  $E_c$  up as  $x$  increases.

Comparing the strained and unstrained band edges in Fig. 15, the effect of the strain is quite pronounced: the strained CBM (VBM) is systematically lower (higher) in energy than its corresponding unstrained value. For the VBM, we even see a change in slope with increasing  $x$  as a consequence of the elastic deformation potentials. As a result, the band gap of  $\text{GaAs}_{0.86}\text{P}_{0.14}$  increases slightly, from 1510 meV for  $x=0.008$  to 1533 meV for  $x=0.2$ , much less than it would be expected from pure chemical offsets only.

### *3. Intradot absorption spectra*

By means of the CI approach described in Ref. 40, we have calculated the intradot absorption spectra for the  $\text{In}_{0.47}\text{Ga}_{0.53}\text{As}/\text{GaAs}_{1-x}\text{P}_x$

potentials, the experiments were carried out at room temperature. Thus, a direct comparison is possible only after applying a temperature correction of  $\approx 70\text{--}100$  meV for the optical gap.<sup>33,51</sup>

The current-voltage measurements presented in Fig. 5 (see Sec. III) show an increase in the open-circuit voltage  $V_{oc}$  when the P composition has been varied from  $x=0.008$  to  $x=0.14$ . The maximum  $V_{oc}$  increase expected from increasing the average band gap of the absorbing region is 0.17 eV, which corresponds to the difference in band gap expected between unstrained  $\text{GaAs}_{0.83}\text{P}_{0.17}$ , 1.71 eV, and  $\text{GaAs}_{0.992}\text{P}_{0.008}$ , 1.54 eV. However, in our strain-balanced QD cells, the  $\text{GaAs}_{1-x}$





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