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Temperature and Bi-concentration dependence of the bandgap and spin-orbit splitting in InGaBiAs/InP semiconductors for mid-infrared applications

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Replacing small amounts of As with Bi in InGaBiAs/InP induces large decreases and increases in the bandgap, E_g , and spin-orbit splitting, Δ_{SO} , respectively. The possibility of achieving $\Delta_{SO} > E_g$ and a reduced temperature (T) dependence for E_g are significant for suppressing recombination losses and improving performance in mid-infrared photonic devices. We measure $E_g(x, T)$ and $\Delta_{SO}(x, T)$ in $\text{In}_{0.53}\text{Ga}_{0.47}\text{Bi}_x\text{As}_{1-x}/\text{InP}$ samples for $0 \leq x \leq 0.039$ by various complementary optical spectroscopic techniques. While we find no clear evidence of a decreased dE_g/dT ($\approx 0.34 \pm 0.06$ meV/K in all samples) we find $\Delta_{SO} > E_g$ for $x > 3.3\text{--}4.3\%$. The predictions of a valence band anti-crossing model agree well with the measurements. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4768532>]

There is increasing interest in incorporating bismuth into conventional III–V semiconductors, due to their potential to optimise the band-structure and overcome several deficiencies in near- and mid-infrared (mid-IR) lasers. Substituting As with small amounts of Bi creates an energy level in the host valence band (VB) causing a VB anti-crossing (VBAC) interaction.¹ This strongly reduces the bandgap E_g by ~ 80 meV/Bi% and ~ 56 meV/Bi% in GaBiAs and InGaBiAs, respectively.² Similar to alloys such as InGaAs, the temperature (T) dependence of the bandgap, dE_g/dT , is expected to be weaker in bismides than in conventional III–Vs. Oe and Asai suggest that, because InBi and GaBi are semi-metallic, InGaBiAs structures with temperature-insensitive E_g may be possible.⁴ Some reports indicate dE_g/dT is significantly weaker in GaBiAs,⁵ but there is controversy by how much,^{6,7} and on the comparative influences of the VBAC interaction and material inhomogeneity. There is relatively little information on dE_g/dT for InGaBiAs; Devenson *et al.* recently reported a *qualitatively* weaker dE_g/dT compared to InGaAs.⁸ This is crucial to understand not only the physics, but also the extent to which device emission wavelength could be stabilised with temperature. Currently, the latter requires thermoelectric cooling, consumes significant power, reduces overall efficiency, and adds complexity and cost. Of further interest is the prediction that adding Bi strongly increases the splitting energy, Δ_{SO} , via an enhanced electron spin-orbital (SO) angular momentum interaction within the heavier Bi atoms. If Δ_{SO} can be made larger than E_g , this offers a route to reduce, or even eliminate, fundamental loss processes such as Auger recombination and inter-valence-band absorption, which dominate IR emitters.³ This could significantly improve the temperature stability of device output power, with the added benefit of growth on well-established relatively inexpensive substrates:

GaBiAs/GaAs is suitable for near-IR, particularly $1.5\text{ }\mu\text{m}$ telecom devices; InGaBiAs could span a wide spectral range from the near-IR out to $>6\text{ }\mu\text{m}$, while still maintaining tolerable strain levels on standard InP substrates. Here, we use experiment and theory to study the key band-structure parameters of InGaBiAs/InP alloys as functions of composition and temperature. We measure optical absorption, photoluminescence (PL), and photo-modulated reflectance (PR) spectra of as-grown InGaBiAs/InP wafers to characterise the influence of Bi on E_g , dE_g/dT , and Δ_{SO} . Specifically, we look for evidence of a possibly reduced dE_g/dT with Bi content, a cross-over between Δ_{SO} and E_g , and the extent to which Δ_{SO} and E_g can be modelled by VBAC theory.

The sample growth was described in Ref. 2. The samples are 260–800 nm thick $\text{In}_{0.53}\text{Ga}_{0.47}\text{Bi}_x\text{As}_{1-x}$ epilayers, with $x = 0, 0.011, 0.024$, and 0.039 , grown by molecular beam epitaxy on a 50–80 nm $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer on standard (100)-oriented InP:Fe substrates.² Nominal In fractions were kept constant throughout at 53% ($\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ is lattice-matched to InP). Bismides require significantly lower growth temperature compared to standard III–Vs; too high and the Bi evaporates, too low and clusters form. These samples were grown at 300–315 °C.² The room temperature (RT) optical absorption spectra were measured with a UV-visible-near-IR spectrophotometer. The absolute absorption coefficient α was calculated from transmission and reflectivity spectra, and the known epilayer thicknesses. For the PL, the samples were excited with a chopped variable power 532 nm diode-pumped solid-state laser (sample spot diameter ~ 1.9 mm) and measured with a grating spectrometer and lock-in amplifier using InGaAs or InSb detectors. PR has several advantages over PL and absorption, e.g., the ability to measure higher-order critical-point energies such as $E_g + \Delta_{SO}$. Its sharp derivative-like features can be fitted with third derivative functional forms (TDFFs) to obtain these

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energies.⁹ For PR and PL, the samples were cooled in a closed-cycle helium cryostat.

Absorption, PL, and PR spectra were observable in all samples with $x \leq 2.4\%$. For the highest, 3.9% Bi, sample, although the InGaBiAs absorption band edge was visible, the PL and PR were weak, apart from PR features due to the InGaAs buffer and InP substrate. Figure 1 shows example PL spectra for 0%, 1.1%, and 2.4% Bi as a function of temperature. We first discuss the 0% Bi sample for which good PL was obtained with 200 mW laser power (sample power density $\sim 7.2 \text{ W/cm}^2$) with peak height decreasing by ~ 28 times between 30 K and 295 K. We might note that, compared to high quality bulk-like epitaxial $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$, the band-to-band PL was weaker and broader, with a full-width at half-maximum (FWHM) of the 30 K peak at 0.786 eV of $\sim 13.2 \text{ meV}$ (as compared to 2.9 meV at 4.2 K (Ref. 10)) which are both possible manifestations of the sample quality due to the lower-temperature growth. However, there was no evidence of deep-level PL,¹¹ indicating reasonable material quality. The 30 K PL also showed peaks at 0.766 eV and 0.734 eV (the latter not discernible in Fig. 1) which we assign to a donor-acceptor transition and its longitudinal optical (LO) phonon replica, respectively, since $\sim 32 \text{ meV}$ corresponds to the LO-phonon energy in InGaAs (using a linear interpolation between the LO-phonon energies of GaAs and InAs (Ref. 12)).

At high temperature, the Bi-containing samples generally gave weaker and noisier PL, requiring higher laser pump powers. The PL intensity decreased with increasing Bi concentration, with the 2.4% Bi sample giving no clear band-to-band PL peak at E_g at any temperature (a very weak and broad band at $\sim 0.6 \text{ eV}$ was seen at $\sim 250 \text{ K}$ only). Furthermore, unlike the

0% Bi sample, the bismide PL showed unusual temperature dependence; Fig. 1(b) shows that the relatively weak band-to-band PL at high temperature disappears below $\sim 160 \text{ K}$, to be replaced by strong long wavelength PL (1.8–2.7 μm) which, even at small excitation powers of 1–2 mW, is >100 times stronger than the band-to-band PL. In the 1.1% Bi sample at 30 K, the long wavelength peak is broad, spanning from $\sim 0.46 \text{ eV}$ (2.7 μm) to 0.65 eV (1.8 μm), centered near $0.55 \pm 0.04 \text{ eV}$ (2.25 μm), and of FWHM $\sim 0.12 \text{ eV}$, while in the 2.4% sample, it is about 3 times weaker, peaking near $0.51 \pm 0.03 \text{ eV}$ (2.43 μm) and of FWHM $\sim 0.07 \text{ eV}$. We do not attribute this peak to any deep-level donor-acceptor recombination, since one would then also expect to see strong band-to-band PL.¹¹ We propose that this fact, together with the rapid drop in the long wavelength PL strength with heating, is explained by compositional inhomogeneity—specifically, the presence of thin Bi-rich areas/layers. Such localised narrower-gap regions would efficiently trap carriers at low temperature producing the long wavelength emission. This is consistent with recent Rutherford backscattering spectrometry of similar samples, which reveals that the Bi concentration increases in the growth direction.¹³

Figure 2 shows the PR spectra of the 1.1% and 2.4% Bi samples at different temperatures. In contrast to the PL, the PR yielded clear features near E_g at all temperatures that strengthened with cooling, as well as higher-energy SO ($E_g + \Delta_{\text{SO}}$ —see Fig. 3), and InP substrate features. However, as Bi content increased from 1.1% to 2.4%, the $\sim 30 \text{ K}$ PR signal dropped by >3 times. Figure 2 shows the fitted E_g energies as open circles (for discussion of PR fitting see Fig. 3). No separate PR features were seen corresponding to the aforementioned localised Bi-rich regions. This suggests that these narrow-gap regions represent only a small fraction of the bismide epilayer, thus giving an insignificant PR contribution, since PR essentially detects the modulation of major critical

FIG. 1. (a) PL spectra of the 0% Bi InGaAs/InP sample at 200 mW laser power at different temperatures. Spectra are normalised to the same peak height (295 K spectrum is ~ 28 times weaker than at 30 K). (b) PL spectra of InGaBiAs/InP samples with 1.1% (solid curves) and 2.4% Bi (dashed curves) at various laser powers and temperatures: 1.1% Bi—50 mW (at 30, 60, 90, and 120 K), 200 mW (160, 190, and 220 K), 300 mW (250 and 290 K), 500 mW and 1 W (both at 290 K); 2.4% Bi—200 mW (30 and 90 K) and 2 W (250 K).

FIG. 2. Example PR spectra of (a) the 1.1% and (b) the 2.4% Bi-containing samples at various temperatures in region of E_g . Open circles indicate fitted transition energy of lowest-energy feature (see also Fig. 3). Feature near 0.73 eV at 296 K is due to $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer layer.

FIG. 3. PR spectrum (symbols) with fitting curves (solid curves), squared absorption coefficient spectrum (dashed curve with linear fit to determine E_g), and PL spectrum at 295 K for InGaBiAs/InP sample with 1.1% Bi. Arrows indicate bandgap determined by each technique. Inset plots absorption coefficient on logarithmic scale.

points in the dielectric function. In contrast, due to the different mechanisms involved in PL, the low-temperature trapping of carriers in these Bi-rich regions results in a relatively efficient emission recombination channel.

Figure 3 compares absorption, PL, and PR spectra for the 1.1% Bi sample at RT. The arrows indicate the extracted E_g values, showing that the three techniques agree well. E_g from absorption is obtained from a conventional linear fit (solid line) of α^2 versus photon energy.¹⁴ For the PL, E_g is taken from the peak position, after subtracting $kT/2$ to account for the effects of thermal carrier distribution.¹⁵ The PR line shapes near E_g are complex and could not be fitted with a single TDFF; it generally required three, as shown by the example curve in Fig. 3. We interpret the highest energy feature (~ 0.73 eV in Fig. 3) as being due to the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ buffer. The remaining two fitted oscillators near the bismide bandgap represent a closely spaced doublet, the lowest-energy component being indicated by the arrow (and, in Fig. 2, by the open circles). A possible interpretation of the central higher-energy component is either the existence of a low-Bi (Bi $\sim 0.6\%$) InGaBiAs layer near the interface with the InGaAs buffer in this thicker epilayer, or heavy-/light-hole splitting due to compressive strain, which may be caused by composition variations of both In and Bi. While this E_g splitting can be seen in the PR, it is not resolvable in the absorption or PL spectra, most likely due to the large broadening and low optical power density. Although the epilayers are relatively thick, there is evidence from recent X-ray diffraction experiments that they are fully strained across their whole thickness.¹³ Even in the Bi-free nominally lattice matched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ reference sample, the PR spectra also resolved a closely spaced doublet near E_g , again suggesting some strain and lattice mismatch in this sample. This may be caused by the lower growth temperature compared to standard $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$.¹³ Future work will study this effect.

Considering first our RT results for E_g and Δ_{SO} , Fig. 4 summarises these for all the samples. Consistent with theoretical predictions, there is clear evidence of a drop in E_g and

FIG. 4. Measured bandgap, E_g , and spin-orbit splitting, Δ_{SO} , from room temperature absorption, PL and PR spectra (symbols). Curves show VBAC calculations for InGaBiAs for different In compositions from 0% to 53%, versus Bi fraction.

a rise in Δ_{SO} with Bi content, with a resonance between them near 3.2% Bi. To model these results we use the VBAC theory¹ for InGaBiAs, using similar methods to those for GaBiAs.¹⁶ Compared to GaBiAs, however, the incorporation of In shifts the valence band maximum (VBM) upwards and increases the separation of the VBM and Bi level, reducing the variation of E_g with Bi%. The relative alignment of the band edges is calculated using model-solid theory.¹⁷ The other quaternary alloy parameters are determined from those of the constituent binary materials.¹⁸ The extra strain effects on the VBM due to the addition of In are also accounted for. Figure 4 shows the resulting theoretical curves for E_g and Δ_{SO} of InGaBiAs/InP samples of varying In compositions, showing good agreement with experiment at RT. For $53\% \geq \text{In} \geq 40\%$, the VBAC model predicts a cross-over between E_g and Δ_{SO} at around 3.3–4.3% Bi. This is also consistent with recent contactless electro-reflectance studies of dilute-Bi InGaBiAs alloys.¹⁹ In materials where $E_g < \Delta_{\text{SO}}$ Auger recombination processes involving excitation of a hot hole into the SO split-off band, and inter-valence band absorption, will both be eliminated, which is predicted to enhance device performance.³ Our measurements indicate this occurs for $E_g \leq 0.55$ eV ($\lambda > 2.2$ μm), showing promise for low Auger and optical losses in mid-IR devices with the convenience of growth on an InP substrate. The PR shows the $(E_g + \Delta_{\text{SO}})$ transition energy as almost static with Bi fraction, suggesting that the Δ_{SO} increase is largely due to the upward movement of the VB edge. However, adding Bi leads to a higher average group V atomic mass, pushing down the SO band. Together, this suggests that the conduction band edge also moves down in energy, implying a type-I conduction and valence band alignment at the InGaBiAs/InGaAs interface, as in GaBiAs/GaAs.²⁰

The inset to Fig. 3 plots α on a semi-log scale showing the onset of weak absorption from below-gap localised states and that $\alpha(E_g) \sim 2500 \text{ cm}^{-1}$. This is similar to the corresponding $\alpha(E_g)$ for our low temperature 0% Bi InGaAs reference sample, implying that Bi does not significantly perturb the host density of extended states. Some evidence for localised states is also observed in the PL data. However, since

FIG. 5. (a) Temperature dependence of PL and PR E_g values for 0, 1.1, and 2.4% Bi. Solid curve is Varshni fit of PL E_g of 0% Bi sample with parameters: $\alpha = 0.50$ meV/K, $\beta = 327$ K, $E_0 = 0.787$ eV. Dashed lines are linear fits of $E_g(T)$ from 150 to 300 K, numbers near each line giving gradients, dE_g/dT (meV/K). Lower dashed-dotted curves show extrapolated long wavelength PL peak energies, using same Varshni fit as for 0% Bi bandgap.

the PL and absorption agree well with PR, a technique insensitive to localisation, we therefore use the conventional definition of extended-state bandgap. Fig. 3 shows all three experimental values of E_g are consistent and agree with the VBAC calculation of the extended-state E_g (Fig. 4).

Figure 5 summarises the temperature dependent measurements of E_g , showing good agreement between the PR and PL results for the different samples. Note that the error in the PL E_g is estimated as half of the FWHM of the corresponding PL peak. The temperature dependence of the $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ PL results for E_g can be fitted well with a Varshni curve,²¹ with fit parameters $\alpha = 0.50$ meV/K, $\beta = 327$ K, and $E_0 = 0.787$ eV. These agree with those of standard lattice matched $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ grown at higher temperature, except for the 0 K bandgap, E_0 , which is slightly less than the accepted value of 0.803 eV.²¹ As discussed earlier, this reduction in E_g may be due to the slight composition and strain variations due to the low-temperature growth. Figure 5 also shows linear fits of $E_g(x, T)$ between 150 K and 300 K. The gradients dE_g/dT show no clear evidence of any reduction with increasing Bi concentration; it has an average value of 0.34 ± 0.06 meV/K. However, due to experimental uncertainties and weak PL in our samples with $> \sim 1.1\%$ Bi, we cannot rule out a slight drop in dE_g/dT with increasing Bi. The aforementioned slight inhomogeneity in these samples may also play a role in the observed dE_g/dT .

The observed photon energies of the strong long wavelength PL peaks are also shown in Fig. 5, with extrapolations to 300 K assuming the same Varshni behaviour as for the 0% Bi sample, giving E_g values of 0.44 ± 0.03 eV and 0.48 ± 0.04 eV for the 2.4% and 1.1% samples at RT, respectively. Examination of the calculated E_g curve versus Bi composition in Fig. 4, for the case of 53% In, indicates that these E_g values correspond to Bi contents of $5.2 \pm 0.6\%$ and $4.4 \pm 0.7\%$, for the suspected thin Bi-rich areas/layers in the 2.4% and 1.1% Bi samples, respectively. This, and the fact that the associated long wavelength PL observed here is

strong, could be consistent with an observed enhancement in PL intensity in optimised GaAsBi/GaAs samples for Bi fractions up to $\sim 4\text{--}6\%$,²² but a more detailed analysis of this is reserved for future studies when $\text{In}_{0.53}\text{Ga}_{0.47}\text{Bi}_x\text{As}_{1-x}/\text{InP}$ samples over a wider set of x become available.

In summary, we studied the fundamental band-structure and optical properties of $\text{In}_{0.53}\text{Ga}_{0.47}\text{Bi}_x\text{As}_{1-x}/\text{InP}$ samples for $x \leq 3.9\%$ from 30–300 K using multiple optical spectroscopic techniques. We especially searched for evidence of the predicted resonance between the bandgap E_g and spin-orbit energies, Δ_{SO} , and for the reduced dE_g/dT suggested recently in the literature. The PR resolved clear E_g and $E_g + \Delta_{\text{SO}}$ features, with $E_g \approx \Delta_{\text{SO}}$ near $x = 3.2\%$ at RT. The increase in Δ_{SO} with Bi fraction is largely due to the upward movement of the VB edge. This result is highly significant with regard to producing low loss mid-IR devices based on InGaBiAs/InP at higher Bi concentrations. Both the PR and PL results yielded similar temperature dependencies for E_g from 150–300 K. However, we find no clear evidence of the suggested reduction in dE_g/dT with increasing Bi content, with a measured average over all samples of $dE_g/dT = 0.34 \pm 0.06$ meV/K. RT measurements of E_g and Δ_{SO} are consistent with valence band anti-crossing.

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