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Optical property comparison of GaAsBi and TlGaAs measured by variable angle spectroscopic ellipsometry

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ABSTRACT

III-V semiconductors have broad uses in optoelectronics due to their direct band gaps and high carrier mobilities. GaAs_(1-x)Bi_x and Tl_xGa_(1-x)As ternary alloys are of interest for light emitting, light absorbing and other applications (e.g. communication lasers, photovoltaics, and high speed transistors) in the infrared spectrum due to their decreased bandgap relative to GaAs. While GaAs has been extensively studied, the optical properties of GaAsBi and TlGaAs are less documented and show significant variation with Bi and Tl content respectively.

This study characterized the optical properties of GaAsBi and TlGaAs films of varying Bi and Tl composition using variable angle spectroscopic ellipsometry (VASE) in a range of temperatures from 25 °C – 300 °C. GaAsBi films were grown between 3.3% and 6.5% bismuth. TlGaAs films were grown between 1.7% and 2.7% thallium. Modeling using a superposition of Gaussian oscillators fit to the dielectric functions of sample layers was used to separate film optical properties from the pseudo-optical properties of the sample.

The analysis in this study directly compares the inclusion of the two largest III-V constituent atoms, Bi and Tl. Comparison of the refractive index and absorption coefficient of samples was done over a spectral range of 0.5 eV to 5 eV (250 nm to 2500 nm). This region displays the absorption edge corresponding to the bandgap of the material, which is then correlated to the incorporation of Bi and Tl in the samples. This characterization allows for better modeling of these alloys for both a fundamental understanding of their properties and for their inclusion in future devices.

Keywords: GaAsBi, TlGaAs, spectroscopic ellipsometry, thermophotovoltaics, direct bandgap, VASE, gallium arsenide, narrow bandgap

1. INTRODUCTION

The need for reliable and efficient energy generation is a strong driving force for today's new photonic technology. In order to meet the demand for better photovoltaic cells, research is being conducted to create and test new III-V alloys which will enable fabrication of devices with better efficiency, higher temperature stability, and a broader spectral range for conversion of light to electricity. For these photonic devices it is desirable to develop materials with tunable, direct bandgaps targeting low energy photons which would extend the spectrum of radiation that can be converted to electrical energy further into the infrared. Such materials will allow for breakthroughs in both thermophotovoltaic (TPV) technology for primary energy generation and waste heat harvesting, as well as small bandgap subjunctions for multijunction solar cells.

Two materials, GaAsBi and TlGaAs, have shown to be promising as low bandgap semiconductors. By varying the composition of Bi and Tl respectively, these materials exhibit significant tunability in the absorption of infrared light. It has been shown that GaAsBi can be used to make long wavelength lasers¹ and diodes for high efficiency photovoltaic cells². TlGaAs may also be viable for similar applications as GaAsBi.

Both GaAsBi and TlGaAs have been relatively unexplored until now due to difficulty in achieving both high incorporation of Bi or Tl while maintaining good material quality. Bismuth and thallium are the largest non-radioactive atoms of groups V and III respectively, with bismuth being the largest non-radioactive atom in general, and because of this they present a significant challenge for epitaxial growth. Due to atomic size mismatch and low miscibility in GaAs, incorporation of Bi and Tl is experimentally hard to achieve. Often incorporation of these larger atoms leads to highly stressed films and surface defects such as metallic droplets. For GaAsBi, due to the nature of the carrier transitions in

these ternary alloys, a small incorporation of Bi atoms will result in a significant change in semiconductor properties³. TlGaAs has been studied less than GaAsBi but also promises interesting optical behavior with increasing Tl content⁴. With advances in growth techniques and material qualities comes the need for more in depth characterization of the optical properties of these materials.

In this study samples of GaAs_(1-x)Bi_x and Tl_xGa_(1-x)As have been grown via molecular beam epitaxy (MBE) over a range of compositions. Optical properties have been measured using variable angle spectroscopic ellipsometry (VASE) to characterize the effect of varying Bi or Tl content. Measurements were taken ranging from room temperature to 300 °C in order to test the shifts in the semiconductor's absorption and stability at higher temperatures. This data is critical information for designing optoelectronic devices suitable for high temperature environments.

2. THEORY

2.1 GaAsBi

Alloys with small bismuth (Bi) incorporations, such as GaAsBi, have been shown to significantly lower the bandgap of their host material (GaAs). This makes them a material of interest for designing low energy optoelectronics. The reduced band gap is attributed to a valence band anticrossing (VBAC) effect where energy states in the GaAs valence band have degeneracy with the energy states of the bismuth impurities⁵. This also causes significant spin-orbit splitting. GaBi has not been successfully grown, however it has a proposed bandgap of -1.45eV. This, and an additional bowing parameter, allows for the calculation of expected GaAsBi band edge as a function of Bi content (x) using the following expression:^{6,7}

$$E_{GaAs_{1-x}Bi_x} = xE_{GaBi} + (1-x)E_{GaAs} - bx(1-x)$$

Here b is a bowing parameter specific to bismuth with dependence on x :

$$b = \frac{\alpha}{(1 + \beta x)}$$

Thus an increase in x will contribute a significant reduction to the energy of the band edge. Values for α and β vary in the literature, for the purposes of this paper values have been taken from Masnadi-Shirazi et. al⁶ and Zayan et. al⁷.

2.2 TlGaAs

Theoretical calculations of the electronic band structure suggest a similar model as for GaAsBi but with a different form for the bowing parameter for TlGaAs. The predicted bandgap of TlAs (-1.04 eV)⁸. Ab initio calculations of the band structure have been done to determine the approximate locations of transitions in the valence and conduction bands⁴. Band edge calculations by Akyuz et al. resulted in the following equation for the bandgap of TlGaAs⁸:

$$E_{Tl_xGa_{1-x}As} = xE_{TlAs} + (1-x)E_{GaAs} - bx(1-x)$$

The proposed bowing parameter is composed of three terms:

$$b = b_{VD} + b_{CE} + b_{SR}$$

In this equation b_{VD} is related to the hydrostatic pressure of the component binary lattices (TlAs and GaAs), b_{CE} is dependent on the bonding between the component lattices, and b_{SR} has to do with structural relaxation. Akyüz et al. report values of b for several Tl concentrations, however the dependence on composition is significantly more complicated than that of GaAsBi.

2.3 Ellipsometry

Spectroscopic ellipsometry allows for accurate measurement of the optical properties of a material. In this study, variable angle spectroscopic ellipsometry (VASE) was chosen to characterize GaAsBi and TlGaAs films because it is capable of detecting differences in film composition, property shifts with temperature, thickness, crystal quality, and other physical parameters of the sample which affects optical response⁹. The ellipsometer shines light of a known polarization state onto the sample, and determines the change in polarization and phase shift of the reflected beam. Raw ellipsometry data gives values of $\tan(\Psi)$ and $\cos(\Delta)$, which are the ratio of s and p polarized light and phase shifts respectively of the reflected beam.

The ratio of reflected polarized light, ρ , in the perpendicular p and s planes can be expressed as:⁹

$$\rho = \frac{R_p}{R_s} = \tan(\Psi)e^{i\Delta}$$

Pseudodielectric constants, ϵ_1 and ϵ_2 , are calculated from the raw ellipsometry data by the following relation:⁹

$$\langle \epsilon_1 \rangle + i \langle \epsilon_2 \rangle = \epsilon_{\text{ambient}} \left[\left(\frac{1 - \rho}{1 + \rho} \right)^2 \sin^2 \Phi \tan^2 \Phi + \sin^2 \Phi \right]$$

where Φ is the angle of incidence. These pseudodielectric constants represent the properties of a two-layer model of sample and ambient. From the pseudodielectric function the following relationships can be used to obtain the real and imaginary parts of the complex index of refraction ($n+ik$) and the absorption coefficient:

$$\begin{aligned} \langle n \rangle &= \left(\frac{\langle \epsilon_1 \rangle + \sqrt{\langle \epsilon_1 \rangle^2 + \langle \epsilon_2 \rangle^2}}{2} \right)^{1/2} \\ \langle k \rangle &= \left(\frac{-\langle \epsilon_1 \rangle + \sqrt{\langle \epsilon_1 \rangle^2 + \langle \epsilon_2 \rangle^2}}{2} \right)^{1/2} \\ \langle a \rangle &= \frac{4\pi \langle k \rangle}{\lambda} \end{aligned}$$

where n is the refractive index, k is the extinction coefficient, and a is the absorption coefficient. Critical points or sharp increases in the absorption coefficient as a function of wavelength correspond to strongly absorbing features due to material structure, such as specific atomic bonds, absorptions, or inter-band energy transitions like carriers being promoted from valence band to conduction band in semiconductors.

In order to extract optical properties of a single layer from the pseudodielectric response of a full multilayer structure, software modeling of the ellipsometry data is required. Absorbing substrate layers, intermediate or buffer layers, oxide coatings and surface defects will all contribute to the pseudodielectric function of the sample. A detailed model of the various layers of the sample must be constructed in order to isolate the optical properties of each material.

3. EXPERIMENT

3.1 Sample Growths

All samples were grown by solid-source molecular beam epitaxy (MBE) on Veeco GENxplor systems. Films were grown on quarter pieces of 2" (001)-oriented epi-ready GaAs substrates. Temperature was monitored by band-edge thermometry using a k-Space BandiT system at all stages during growth. Reflection high energy electron diffraction (RHEED) patterns were recorded to confirm native oxide desorption as well as monitor crystalline quality of epilayers. Epi-ready wafers were heated to 620 °C under constant As₄ flux and held until maximum brightness in the RHEED pattern was achieved to desorb their surface oxide. Subsequently GaAs buffer layers were grown to 500 nm thick at 580 °C and at a rate of 0.8-1 monolayers/second (ML/s) measured by RHEED intensity oscillations of the (2×4) surface reconstruction.

After GaAs buffer growth, samples for GaAsBi growth were cooled at 30 °C/min to 250 °C and the As₄ valve was adjusted to achieve an As₄/Ga beam equivalent pressure (BEP) ratio of 12.5. Under As₄ flux only, the RHEED surface reconstruction was c(4×4). GaAsBi growth was initiated by opening the Ga and Bi shutters and assuming a growth rate of 0.45-0.5 ML/s as measured by RHEED oscillations for GaAs. The Bi/Ga BEP ratio ranged from 0.16-0.35 and was adjusted to achieve GaAs_{1-x}Bi_x compositions 0.033<x<0.065. Under Ga, Bi and As₄ fluxes, the RHEED surface reconstruction was (1×3).

After GaAs buffer growth, samples for TlGaAs growth were cooled at 30 °C/min to 220-210 °C and the As₄ valve was adjusted to achieve an As₄/Ga beam equivalent pressure (BEP) ratio of ~5. Under As₄ flux only, the RHEED surface reconstruction was c(4×4). TlGaAs growth was initiated by opening the Ga and Tl shutters and assuming a growth rate of 0.55 ML/s as measured by RHEED oscillations for GaAs. The Tl/Ga BEP ratio ranged from 0.046-0.076 and was adjusted to achieve Tl_xGaAs_{1-x} compositions 0.017<x<0.027. Under Ga, Tl and As₄ fluxes, the RHEED surface reconstruction was difficult to resolve but appeared to be a (1×2).

3.2 Previous Characterization

After growth, samples were characterized by high-resolution x-ray diffraction (XRD) using 2θ-Ω line scans and (224) reciprocal space maps to determine bismuth or thallium composition and strain state. Modeling of GaAsBi in the Bruker

Leptos software assumes a GaBi lattice constant of 0.633 nm and the elastic constants of GaAs¹⁰. Modeling of TlGaAs assumed a TlAs lattice constant of 0.618 nm and elastic constants reported by Beneyton et al¹¹. Composition in select samples was verified by Rutherford backscattering spectrometry (RBS). XRD and TEM were used to independently determine film thicknesses for use in VASE modeling software. Optical microscopy using a Leica differential interference contrast microscope was utilized to confirm the presence/absence of surface droplets.

3.3 Room Temperature Measurement at Varying Angles

VASE scans were taken on all samples in a range of photon energies from 0.5-5 eV using a J.A. Woollam Co. VASE® ellipsometer. This energy range was chosen to display the primary band edge as well as several of the higher energy transitions above the band gap. This spectral range covers the first 4 energy transitions in GaAs, that is the E_g , $E_g + \Delta_{so}$, E_1 and $E_1 + \Delta_1$ transitions¹². Data was collected at room temperature in ambient atmosphere. The angle of incidence chosen was between 70° and 80° to collect data around the pseudobrewster's angle of the samples. This was done to maintain a value of Δ around 90 degrees, which improved sensitivity and increased accuracy of the ellipsometric data¹². In order to reduce signal noise the software performed dynamic averaging whereby it adjusted the number of analyzer cycles to cut the average noise to a minimum¹³. Using this method it was possible to create cleaner data near the absorption edge, which allowed for more accurate calculation of the band gap energy.

3.4 Modeling

Modeling of sample and film optical properties was done using J.A. Woollam's WVASE software for windows. Ellipsometry data was analyzed using a model of 4 layers including a GaAs substrate, GaAs buffer layer, GaAsBi or TlGaAs layer, and an appropriate thin oxide layer. A point by point fit of the data was utilized in order to isolate the optical properties of the material of interest from those of the bulk sample. Once the layer of interest was isolated in the software it was modeled using a set of parametrized Gaussian oscillators fit to the critical points of the spectrum, and the software varied the parameters in order to fit the data with the lowest mean squared error (MSE) value. A parametrized semiconductor oscillator (Psemi-M0) was used to fit the band edge as it provided a sharper tail to the absorption characteristic of direct bandgap semiconductors. Once the modeling process was complete and a low MSE was achieved the optical properties of the material of interest could be examined alone. Low MSE values equal to or less than 2.6 were achieved for each GaAsBi sample and less than 3.8 for each TlGaAs sample, which confirmed the accuracy of the modeled values.

3.5 Measuring GaAsBi and TlGaAs bandgaps

Bandgaps were calculated for each sample from plotting the absorption coefficient squared vs. photon energy. In direct bandgap semiconductors, the absorption coefficient squared shows a linear trend directly above the band edge. The band edge can then be measured by extrapolating a linear least-squares fit down to the energy axis.⁶ When calculating the linear regression around the band edge only the region above the exponential Urbach tail was taken into consideration. The Urbach tail in GaAsBi is the product of crystalline lattice quality, and the addition of bismuth in the range of concentrations presented here significantly increases the number of below bandgap states.⁶ A significant below band-edge tail was also seen for the TlGaAs samples.

3.6 Elevated Temperature Measurements

For temperature measurements a Linkam Scientific Instruments heating stage with CaF₂ windows was fitted to the ellipsometer and was used to heat the sample up to 300 °C under a nitrogen or helium purge gas flow. Because of the construction of the heating stage measurements were taken at an incident angle of 70 degrees. Samples were not tested higher than 300 °C due to the risk of desorption of the constituent atoms which would irreversibly alter the composition of the sample and make the optical properties invalid past that point.

4. RESULTS AND DISCUSSION

4.1 GaAsBi Optical Properties

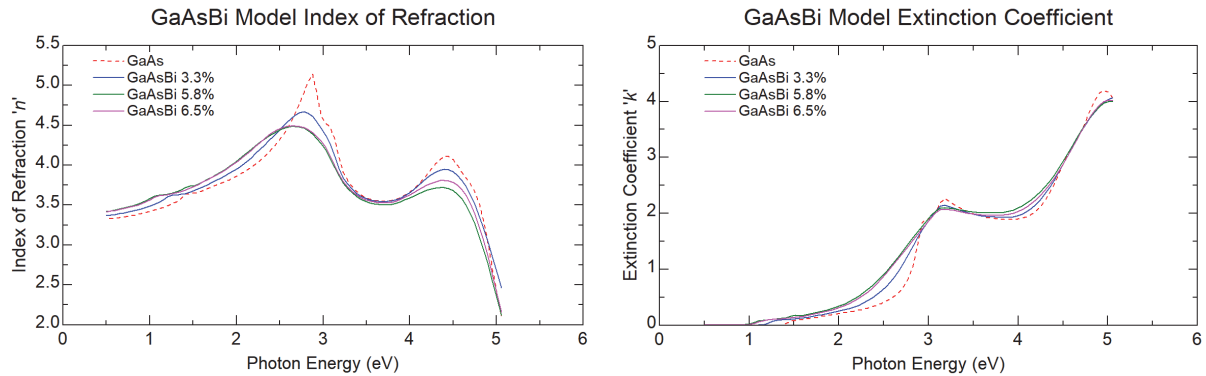


Fig.1 Modeled GaAsBi optical properties n (left) and k (right) vs. photon energies for samples ranging from 3.3% Bi to 6.5% Bi. The optical properties of GaAs sample are shown by the red dotted line for reference.

With increasing Bi content the GaAsBi samples showed dramatic shifts towards lower energy absorption as predicted by theory (Fig. 1). The region between 2 and 3 eV shows good correlation between increased Bi and lower band-edge energy absorption. The above gap energy transitions in the range around 3 eV become less pronounced with increasing Bismuth incorporation, with a shift towards lower energy as well. This broadening around 3 eV has been reported in the literature as a change in spin-orbit splitting of the energy transition.¹² The k spectrum also indicates an increase in absorption in the range from 3.5 eV to 4.5 eV and broadening of the absorption feature at ~5 eV.

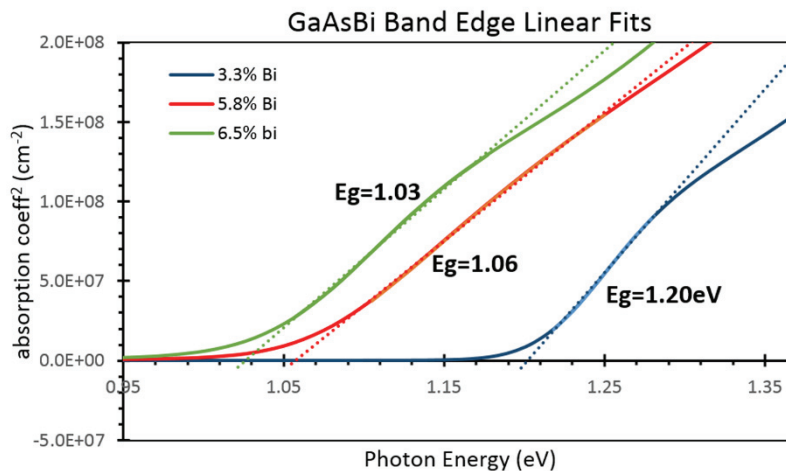


Fig.2 GaAsBi Band edge absorption coefficient (squared) vs. energy for varying Bi samples. Absorption coefficient is squared so that a linear fit to the region above the Urbach tail displays the band gap energy as its x-intercept.

Linear fits to the squared absorption coefficient just above the band edge show the band gap energy of the samples (Fig. 2). As predicted, the band edge moves to lower energy with increasing bismuth content. The bandgaps predicted by theory and determined by experiment and optical modeling are displayed in Table 1. The table shows good agreement between theoretical prediction and the results measured here, with the Masnadi-Shirazi prediction being slightly closer to the measured result. This is likely due to the compressive strain experienced by the GaAsBi layer from the GaAs substrate. Density functional theory results presented in Zayan et al.⁷ are computed for free-standing GaAsBi.

Table 1. Bismuth percentage for each sample, bandgaps reported by Masnadi-Shirazi et al.⁶, DFT predicted bandgap from Zayan et al.⁷, and measured bandgap from this work

%Bi	Masnadi-Shirazi et. al (eV)	Zayan et. al (eV)	Eg measured (eV)
3.3	1.19	1.11	1.20
5.8	1.03	.94	1.06
6.5	0.99	.9	1.03

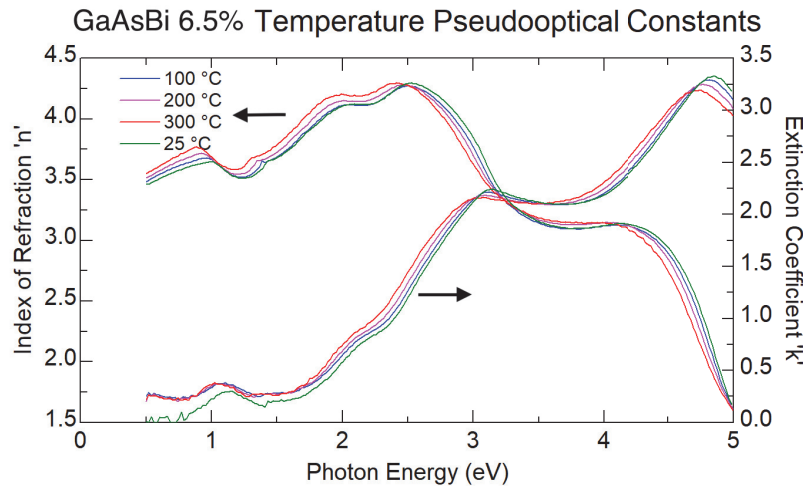


Fig. 3 Pseudo-optical constants for the 6.5% Bi sample at varying temperatures.

Ellipsometric measurements taken at increasing sample temperature resulted in a consistent lowering of the energy for each transition in the optical spectrum of GaAsBi, with pseudo-optical data from the 6.5% Bi sample shown in Fig. 3. Some reduction of feature height is visible in the extinction coefficient peak around 3 eV as well as the peak at 4.75 eV. The peak around 1 eV to 1.2 eV is likely due to optical interference between film layers in the sample. Modeling of the material is required to further explore the optical properties as a function of temperature, especially around the band edge. This work is presently underway.

4.2 TiGaAs Optical Properties

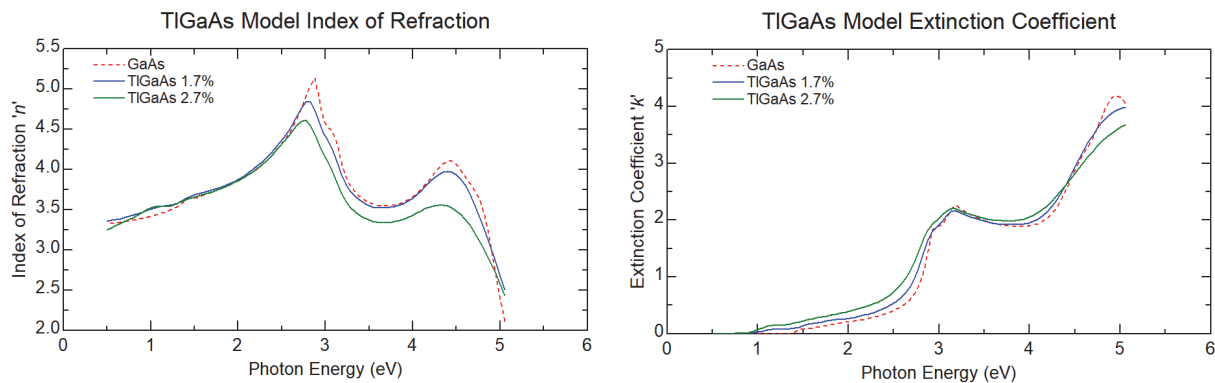


Fig.4 Modeled TiGaAs optical properties n (left) and k (right) vs. photon energy for samples ranging from 1.7% Bi to 2.7% Ti. The optical properties of GaAs are shown by the red dotted line for reference.

Optical property measurements of films with increasing thallium content also show shifts in absorption features towards lower energies (Fig. 4). The extinction coefficient above the band edge between 1 eV and 3 eV shows a consistent shift to lower energies similarly to GaAsBi. The transitions around 3 eV visible in the k data do not decrease and broaden as significantly as they do in the GaAsBi samples, and much of the overall shape of the GaAs transitions is maintained. This indicates the 3 eV transition and its neighboring spin-orbit couple transition are less affected by increasing Tl than increasing Bi. The opposite is true for the higher energy transition at ~ 5 eV which shows a larger decrease and flattening in both n and k when compared to GaAsBi. Increasing scan range to higher energies would allow for further analysis of these transitions. In the n curve, the drop and broadening in the peak at around 2.75 eV is less in TlGaAs than seen in GaAsBi for equal amounts of Tl and Bi respectively. The TlGaAs refractive index trend in the region around 3.5 eV to 4 eV is interesting as it shows a large decrease in n for the higher Tl sample, but the effect is not as pronounced in the lower Tl sample.

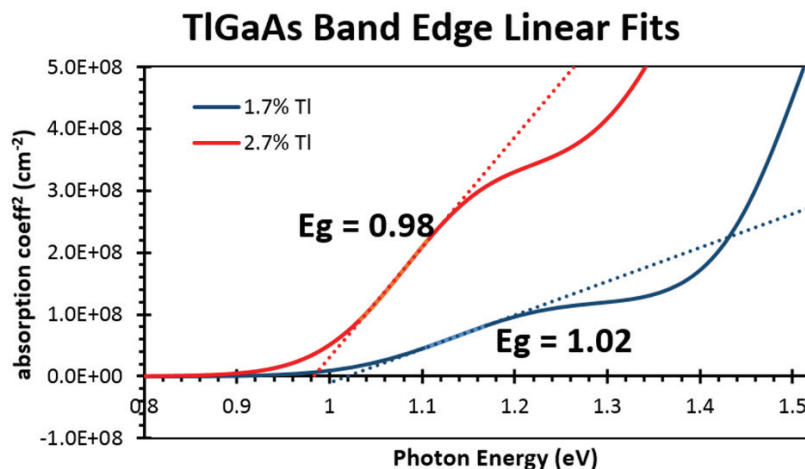


Fig.5 TlGaAs band edge absorption coefficient (squared) vs. energy for varying Tl samples. Absorption coefficient is squared so that a linear fit to the region above the Urbach tail displays the band gap energy as its x-intercept.

Linear fits to the squared absorption coefficient just above the band edge for the TlGaAs samples resulted in the band-edge values shown in Fig. 5. These show on average lower values than were obtained from GaAsBi for similar amounts of Bi added. This shows that incorporation of thallium has a larger effect on the band edge than the addition of similar amounts of bismuth, per percent incorporation.

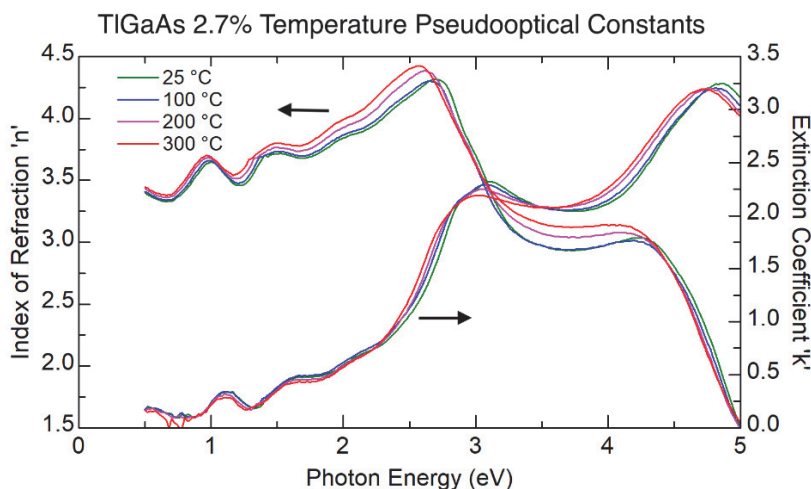


Fig. 6 Pseudo-optical constants for the 2.7% Tl sample at varying temperatures.

Ellipsometric data taken at elevated temperatures shows a drift in TlGaAs transition energies towards lower values with increasing temperature (Fig. 6), similar to what was observed for GaAsBi. The area above the band edge from 1.25 eV to 2.5 eV shows less change with increasing temperatures than optical properties further above the band-edge, which may indicate stability of the material's band edge as a function of temperature. Like with the GaAsBi sample, interference fringes around 0.5 eV to 1.5 eV make determining the exact energy of the band gap difficult. Further modeling is needed to make quantitative claims regarding the relationship between band gap and temperature.

5. CONCLUSIONS

This study provided optical data on two ternary semiconductor compounds, GaAsBi and TlGaAs, both of which have been proposed as low bandgap materials for infrared applications. Theoretical work has shown these materials to be of interest for a variety of photonic applications, and as such thorough characterization using techniques such as VASE is vital for determining the properties of these materials. VASE data was taken over a 0.5-5eV spectral range in which primary band edges as well as higher energy transitions are visible. Data was taken for GaAsBi and TlGaAs samples with a range of Bi and Tl compositions to understand how their optical properties shift with increasing incorporation of their respective third alloying element. Temperature measurements from 25 °C to 300 °C were also made to understand each material's optical behavior in higher temperature environments. Comparison of the band edges of the two materials indicated a larger decrease in bandgap per percentage incorporation of Tl as compared to Bi. Future work hopes to make more quantitative modeling and analysis of relevant energy transitions and other features present in the optical spectrum, as well as modeling the temperature dependence of the primary band edge and other optical features.

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