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Analytical Expressions for the Luminescence of Dilute Quaternary InAs(N,Sb) Semiconductors

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Abstract. In this paper, we calculate the luminescence of the dilute quaternary InAs(N,Sb). The incorporation of N leads to a reduction of the energy gap of the host InAs and Sb acts as a surfactant, improves the N incorporation and further reduces the bandgap. This is thus extremely relevant for devices operating in the mid-infrared (MIR) spectral range from 3 to 5 μm . In order to describe this system, the theory starts with the band anticrossing model applied to both conduction and the valence band to generate inputs for analytical approximations that lead to luminescence spectra, including plasma screening, bandgap renormalization and excitonic enhancements. Direct application of the equations leads to good agreement with some recent experimental data.

Keywords: photoluminescence, dilute semiconductors, many body effects

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1 Introduction

Photoluminescence, or more generally, photon emission due to different excitation processes [1, 2] is a very powerful tool to characterize semiconductor materials and map specific characteristics of new devices [3, 4], notably dilute semiconductor materials, which have strong potential for light-harvesting devices and one of the most widely studied dilute semiconductors is InAsN [5-8]. Similar to other III-V dilute semiconductors, InAsN exhibits a very large energy band gap bowing which is due to the remarkable variations in the atomic sizes and electronegativity [9] between the nitrogen and InAs. This results in the substitution of a small fraction of the N atoms for InAs and hence the conduction band is perturbed by the N-impurity localized state which is usually below the conduction band edge of InAs. The mechanism of the resulting conduction band edge reduction has been explained by the so called conduction band anticrossing model [10,11]. A further improvement has been reported [6,7] on the spectral

qualities of InAsN by the introduction of Sb ion. This yields a narrower band gap InNAsSb alloy semiconductor, and has been successfully done using Sb flux in molecular beam epitaxy and a redshift in band gap on the incorporation of Sb has been reported [9]. Furthermore, band gap engineering by incorporating Sb ion is known to deliver emission at the spectral range between 3 and 5 μm . Thus, devices based on these alloys are potential candidates in medical diagnostics, free space communication and atmospheric pollution sensors [12,13]. Also, strained InAsNSb epilayers on InAs substrate for LED applications have been reported [6]. Just as the presence of the N impurity reduces the conduction band edge, the incorporation of Sb increases the valence band edge thereby causing a further reduction in the band gap [12]. In fact it has been noted that the roles of Sb in InAsN are to serve as a surfactant [6,9] and also that when a little amount is incorporated into the lattice it can be used to tune the band gap of InAs(N,Sb) [6]. Hence InNAsSb alloys have huge potential for the development of optoelectronic devices operating in mid-infrared (MIR) spectral range from 3 to 5 μm , to be used in a large number of applications including medical diagnostics, atmospheric pollution monitoring, and free-space communication. The more complex band anticrossing model required to describe the bandstructure of InAs(N,Sb) where both conduction and valence can be engineered and tailored to specific applications must then be included in an approach such of that found in simpler systems, such as in Ref. [14]. This is achieved in this paper, which is organized as follows: We start with a description of the band structure solution and show how it is included in the Luminescence solver. Next, direct comparisons with experiments in the literature are used to validate the model for different quaternary concentrations. The combined role of plasma screening, bandgap renormalization and excitonic enhancement are put in evidence by comparing and contrasting calculations including these many body effects and the experiments against the free carrier case. These effects have

been included, because they are sufficient to explain the experimental data, without introducing extra momentum dependence that would prevent the development of simple analytical solutions, which are a target for an approach that can be easily used by a large number of colleagues. A brief summary follows.

2 Outline of the mathematical model and material properties

2.1 Bandstructure

The incorporation of *Sb* in *InAsN* semiconductor yields an alloy of the form $InAs_{1-x-y}N_xSb_y$. Here x and y are *N* and *Sb* mole fractions respectively. In the treatment of the band gap energy of $InAs_{1-x-y}N_xSb_y$ alloy, the band anticrossing model is applied on the conduction band and the valence band since the reduction of the band gap energy is due to the presence of both *N* and *Sb* impurities. At the conduction band, *N* is substituted in small amount for the *As* – atom in the binary *InAs* introducing an *N* – isoelectronic level. A similar approach has been applied in the study of *GaAsNSb* [9]. The solutions of the resulting dispersive eigenstates give doubly degenerate subbands described by the conduction band anticrossing model (BAC)

$$E_{\pm}^C(k) = \frac{1}{2} \left[E^N + E_{C,InAs}(k) \pm \sqrt{[E^N - E_{C,InAs}(k)]^2 + 4V_N^2 x} \right], \quad (1)$$

Here E^N , $E_{C,InAs}$ and V_N are the *N* – isoelectronic level, conduction band edge energy and the matrix element describing the coupling constant between the *N* – impurity level and the extended conduction band state. Similarly, in the valence band, the presence of the *Sb* atom introduces an impurity state below the valence band maximum of *InAs*. This supports the application of the result of a 12 x 12 band $\mathbf{k}\cdot\mathbf{p}$ method [15] for valence band anticrossing model with the subband energy eigenvalues

$$E_{\pm}^{hh}(k) = \frac{1}{2} \left[E^{Sb} + H \pm \sqrt{[E^{Sb} - H]^2 + 4V_{Sb}^2 y} \right], \quad (2)$$

$$E_{\pm}^{lh}(k) = \frac{1}{2} \left[E^{Sb} + L \pm \sqrt{[E^{Sb} - L]^2 + 4V_{Sb}^2 y} \right], \quad (3)$$

$$E_{\pm}^{SO}(k) = \frac{1}{2} \left[E^{Sb-SO} + S \pm \sqrt{[E^{Sb-SO} - S]^2 + 4V_{Sb}^2 y} \right], \quad (4)$$

where H and L , denote the valence band energies for the heavy-holes (hh) and light hole (lh) band respectively band of the binary $InAs$ semiconductor. The spin orbit split-off energy (SO) is denoted by S . Within the 12×12 $\mathbf{k} \cdot \mathbf{p}$ band the energies are given as, $H = -\hbar^2 k^2 / 2m_{hh} + \Delta E_{VBM} y$, $L = -\hbar^2 k^2 / 2m_{lh} + \Delta E_{VBM} y$ and $S = (H + L) - \Delta_0 - \Delta E_{SO} y$ respectively, where Δ_0 , m_{lh} and m_{hh} are the spin orbit, light-hole and heavy-hole effective masses of $InAs$ semiconductor. The impurity levels of the heavy holes and the spin orbit split-off bands are denoted by E^{Sb} and E^{Sb-SO} respectively. The matrix element for the coupling constant which describes the interaction between Sb and the valence band states is denoted by V_{Sb} . The theoretical value of the band gap of the quaternary $InAs_{1-x-y}N_xSb_y$ alloy can thus be obtained taking into account the virtual crystal approximation (VCA), as the difference between the band extrema E^c and E^v . In this case the band extrema are given as $E^c = E_-^c - \Delta E_C$ and $E^v = E_+^{hh} - \Delta E_V$, where ΔE_C and ΔE_V are the conduction and valence band discontinuities between the end point binaries i.e. $InAs$ and $InSb$. The valence band positions $E_{v,InAs}$ and $E_{v,InSb}$ of the binaries $InAs$ and $InSb$ can be calculated [16] by using $E_{v,InAs} = E_{vav,InAs} + \Delta_{0,InAs} / 3$ and $E_{v,InSb} = E_{vav,InSb} + \Delta_{0,InSb} / 3$. Their corresponding conduction band positions are $E_{c,InAs} = E_{v,InAs} + E_{g,InAs}$ and $E_{c,InSb} = E_{v,InSb} + E_{g,InSb}$ respectively. The spin orbit split-off energy position are calculated $E_{SO,InAs} = E_{vav,InAs} - 2\Delta_{0,InAs} / 3$ and $E_{SO,InSb} = E_{vav,InSb} - 2\Delta_{0,InSb} / 3$. Here

E_{vav} and Δ_0 represent the average valence subband and spin orbit energy of *InAs* or *InSb* semiconductor respectively. The conduction, valence and spin orbit split-off band discontinuities between the binaries *InAs* and *InSb* are then respectively evaluated as $\Delta E_C = E_{c,\text{InAs}} - E_{c,\text{InSb}}$, $\Delta E_v = E_{v,\text{InSb}} - E_{v,\text{InAs}}$ and $\Delta E_{SO} = E_{SO,\text{InSb}} - E_{SO,\text{InAs}}$.

2.2 Luminescence

The optical response of semiconductor materials can be obtained by self-consistent evaluation of Many body Nonequilibrium Green's Functions (NEGF), which have been successfully applied to intersubband [11,17,18] and interband transitions [19,20] in quantum wells and superlattices. This paper starts from an approach that can also describe superlattices as effective 3D anisotropic media [21] and leads to very accurate approximations. Here, the bandstructure relations described in Sec. 1.1 are inserted in the analytical solution for semiconductor luminescence, which has been derived in Ref. [14] by means of a step by step set of approximations applied to the equation for the interband polarization, within the context of a microscopic Nonequilibrium Green's Functions formalism,

$$I(\omega) = I_0 \left[\frac{\hbar\omega}{E_0} \right]^3 \frac{1}{e^{\beta(\hbar\omega - \mu)} + 1} \left\{ \sum_{l=1}^{g/2} \frac{4\pi}{l} \left[\frac{1}{l^2} - \frac{l^2}{g^2} \right] \delta_{\Gamma} \left(\Delta - \frac{E_l}{E_0} \right) + 2\pi \int_0^{\infty} d\phi \frac{\sinh \pi g \sqrt{\phi}}{\cosh \pi g \sqrt{\phi} - \cosh \pi \sqrt{\phi g^2 - 4g}} \delta_{\Gamma}(\Delta - \phi) \right\}, \quad (5)$$

where, here $I_0 = |\wp|^2 E_0^2 n_b / \pi^2 \hbar^3 c^3 a_0^3$, \wp , n_b , c , E_0 and a_0 are the dipole matrix element, background refractive index, speed of light in vacuum, exciton binding energy and Bohr radius respectively. ϕ , β and μ are the integration variable, inverse thermal energy and quasiparticle chemical potential with respect to the band gap energy of the dilute nitride material E_g . The probe photon energy $\hbar\omega$ is detuned by $\Delta = \hbar\omega - E_g/E_0$, where E_g is renormalized, following the standard Mott criterion as reported in Ref. [21]. The bound state energies are given as

$E_l = -E_0(l^{-1} - g^{-1}l)^2$. The integral runs from $l = 1$ through the square root of the band state factor g . The optical properties depend largely on the band gap of the quaternary alloy which is influenced by Sb and N mole fractions. In fact, one would expect a much stronger dependence on N mole ratio than Sb mole since it's only a small part of the latter that enters the lattice of the quaternary alloy [8]. The distribution of the N mole fraction x , is expected to cause an inhomogeneous broadening of the photoluminescence spectra of $InAs_{1-x-y}N_xSb_y$ alloy. Here, the inhomogeneously broadened luminescence is approximated as a statistical average of the homogeneously broadened luminescence spectra

$$I_{inh}(\omega) = \int_{-\infty}^{\infty} I(\omega, x)G(x)dx, \quad (6)$$

Here, $G(x)$ is the Gaussian linewidth having a nominal N - mole fraction x_0 and standard deviation σ_x given by $G(x) = 1/\sqrt{2\pi}\sigma_x e^{-((x-x_0)/2\sigma_x^2)}$. Details of all expressions above are given in Ref. [14]. The goal of this paper is to extend the approach to quaternary materials and describe corresponding experimental results. In the simulations presented next we just considered a distribution $G(x)$ (Nitrogen related) and let y (Sb) as a fixed parameter.

3 Numerical results and discussions

In our calculations, the N impurity level is taken with the form $E^N = E^{N0} - \alpha x$, where the value $E^{N0} = 1.48eV$ [9], instead of the tight binding approximation values of $E^{N0} = 1.36eV$ in Ref. [5], using $\alpha = 2$ from Ref. [5, 9]. The free fitting parameter V_N is taken as $V_N = 2eV$, following Ref. [22]. The position of the Sb level $E^{Sb} = 1.0eV$ [10] the corresponding spin orbit splitting energy impurity level, $E^{Sb-S0} = 1.6eV$ and the free parameter for the valence band $V_{Sb} = 1.05eV$,

were used in the calculations [10,23]. Figure 1 shows the schematic diagram of the band lineup of $InAs_{1-x-y}N_xSb_y$ using the BAC model.

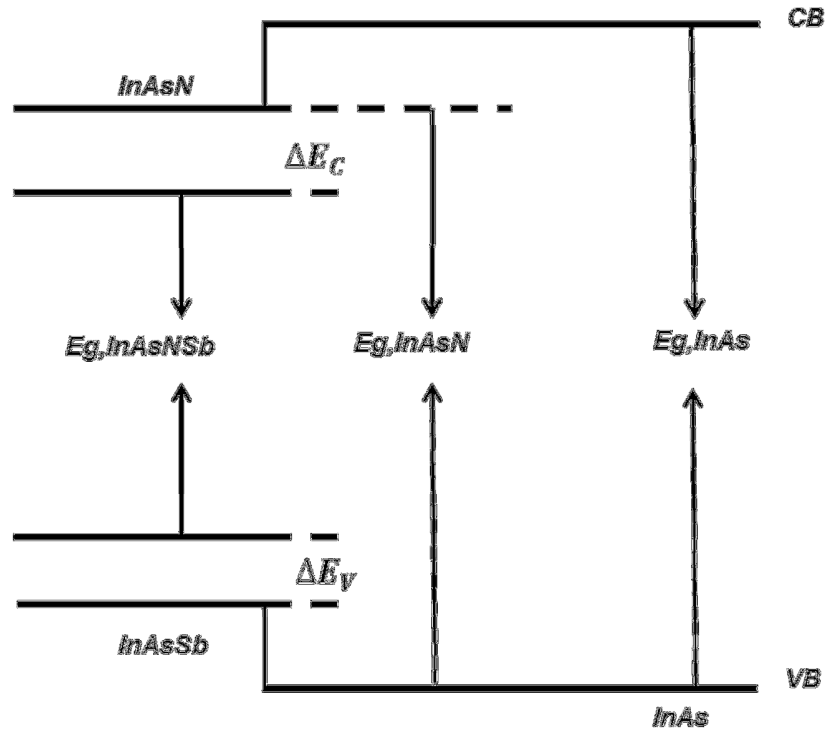


Fig. 1 Band schematics of $InAs_{1-x-y}N_xSb_y$ dilute semiconductors.

The conduction band (CB) minimum is controlled by the N – mole fractions and the valence band (VB) maximum is controlled by the Sb – mole fraction. The Virtual crystal approximation is used to introduce the small band offsets ΔE_C and ΔE_V between the constituent $InAs$ and $InSb$. At zero doping level, i.e. $y = 0$ and $x = 0$, the band gap of the semiconductor is simply $E_{g,InAs}$ as depicted in the diagram above. The material is completely $InAsN$ when the Sb mole fraction is $y = 0$ and the N – mole fraction is increased to a small amount x . In this case the band offsets are zero and the valence band maximum will be that of $InAs$, instead of $InAsSb$ as

can be seen in the diagram. A narrower band gap $InAs_{1-x-y}N_xSb_y$ is obtained when both Sb and N – impurities are present. Figures 2(a) and 2(b) below depict the band structure of $InAs_{1-x-y}N_xSb_y$ with a lattice temperature of 295 K, calculated for the mole fraction combinations (a) 1.4% N , 4.1% Sb and 1.8% N and 7.3% Sb . The plots show the dispersions as depicted in Eqs. (1) to (3) above. The dotted lines show the dispersions of the host binary semiconductor, while the solid lines show the corresponding dispersions of dilute quaternary semiconductor. The relevance of our approach compared to Bose-Einstein ($E_{g, BE}$) and Varshni ($E_{g, Var}$) fits to the bandgap are made clear later in the text when we directly compare free carrier approximations, many body corrections and the fits themselves. These fits are characterized by the following parameters and expressions

$$E_{g, Var} = E_{g0} - \frac{\alpha T^2}{T + \beta}, \quad E_{g, BE} = E_{g0, BE} - \frac{2a_B}{\exp\left(\frac{\theta_B}{T}\right) - 1} . \quad (7)$$

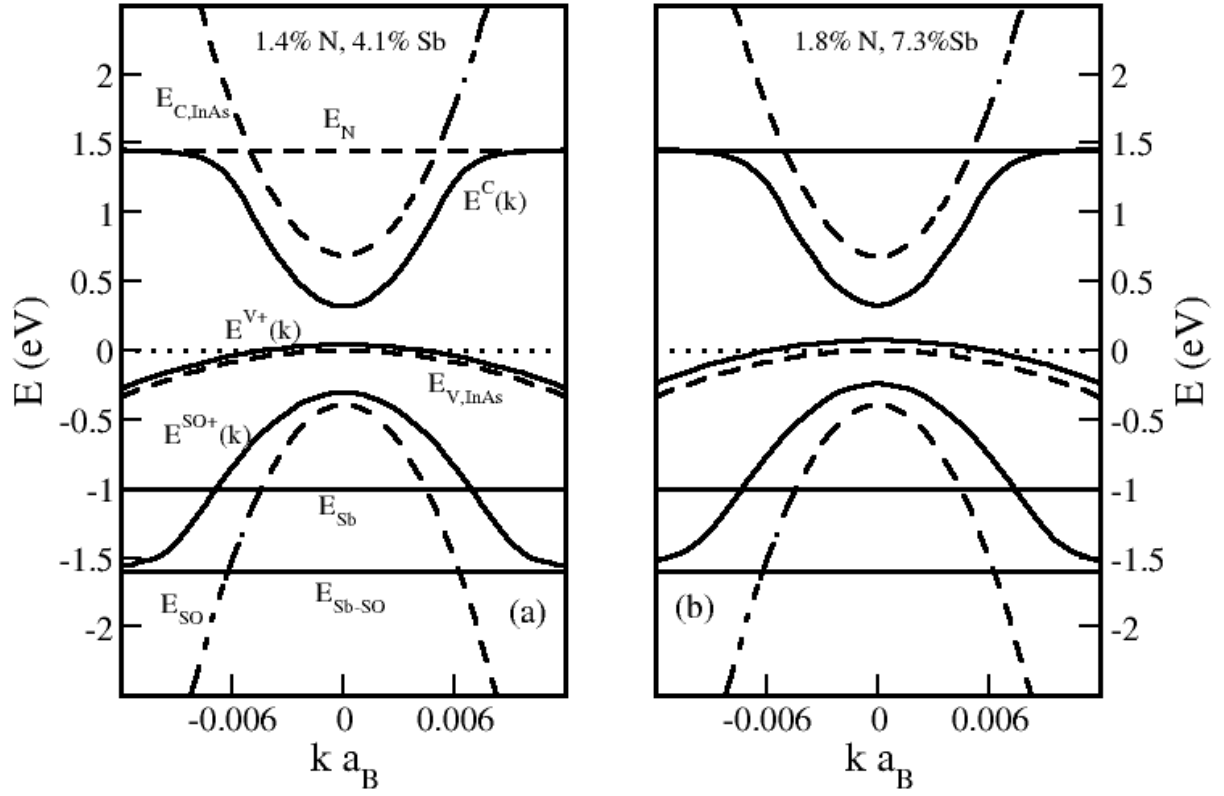


Fig. 2 Band structure of $\text{InAs}_{1-x-y}\text{N}_x\text{Sb}_y$. (a) 1.4% N , 4.1% Sb and (b) 1.8% N , 7.3% Sb . The incorporation of Sb promotes a further reduction in bandgap, although the prominent shrinkage is due to N .

Figure 3 compares the computed and experimentally measured luminescence for the different mole fractions.

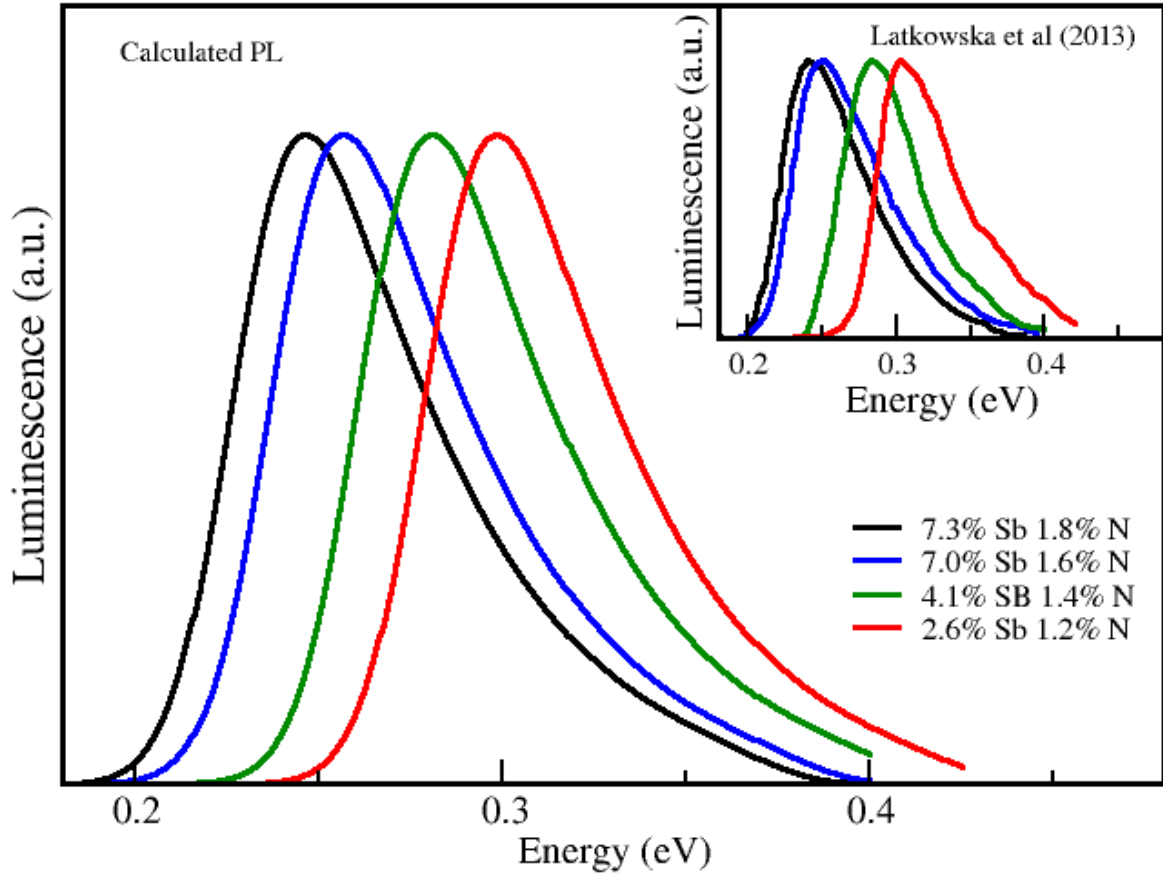


Fig. 3 Room Temperature Luminescence of $InAs_{1-x-y}N_xSb_y$ dilute semiconductor. The plots are the calculated luminescence spectra and the inset plots are the measured luminescence extracted from Ref. [5]. The lines are (from left to right) : 1.8% N , 7.3% Sb (blue); 1.6% N , 7.0% Sb (green); 1.4% N , 41% Sb (red); 1.2% N , 2.6% Sb (cyan). The theory predicts the general feature of a strongly asymmetric spectral shape at room temperature in good agreement with experiments.

Using the inhomogeneously broadened luminescence given in Eq.6 above, a simple Gaussian distribution with standard deviation $\sigma_x = 0.0025$ was sufficient to obtain a spectrum with a reasonable linewidth. All through the calculations, the density used was $n = 10^{14} \text{ carriers/cm}^3$. The calculated luminescence spectra are in good agreement with the

reported experimental results in Ref. [5]. Figure 4 shows the luminescence spectra at different carrier temperatures for two different sample compositions.

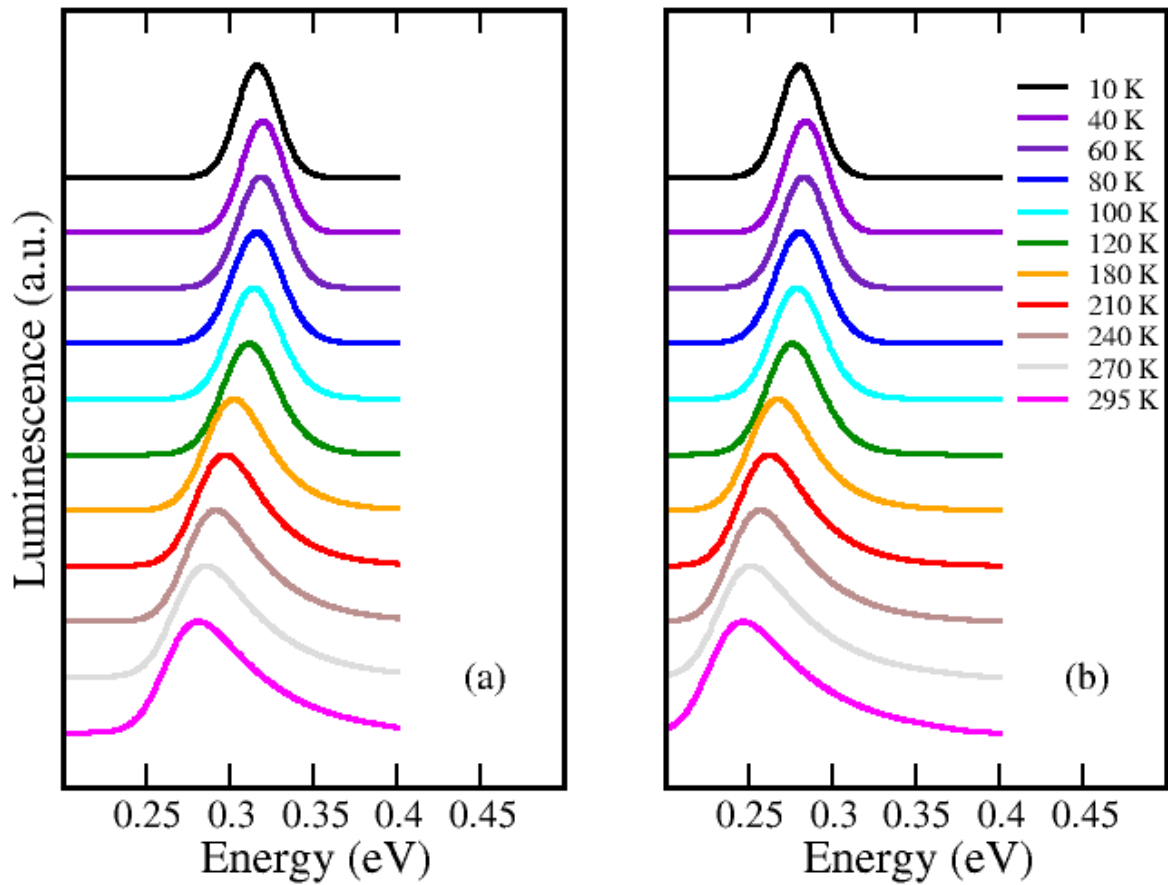


Fig. 4 Calculated luminescence of $InAs_{1-x-y}N_xSb_y$. (a) 1.4% N , 4.1% Sb and (b) 1.8% N , 7.3% Sb for different temperatures increasing from 10 K to 295K. The evolution from a Gaussian-like shape at low temperature to an asymmetric shape at high temperature is a general feature in excellent agreement with the experimental spectra reported in Fig.2 of Ref. [5].

The required dependence of the free carrier energy band gaps with temperature are given by the Varshni relation for the host binary material *InAs* given in Ref. [24], where E_{g_0} is the low temperature band gap of binary *InAs* $E_{g_0} = 417 \text{ meV}$, α and β are the Varshni parameters given by $\alpha = 0.276 \text{ meV} / \text{K}^2$, $\beta = 93 \text{ K}$ (see Eq. 7). In order to highlight the relevance of our approach, Fig. 5 depicts the luminescence energy peaks with respect to temperature for the sample with 1.4% N and 4.1% Sb.

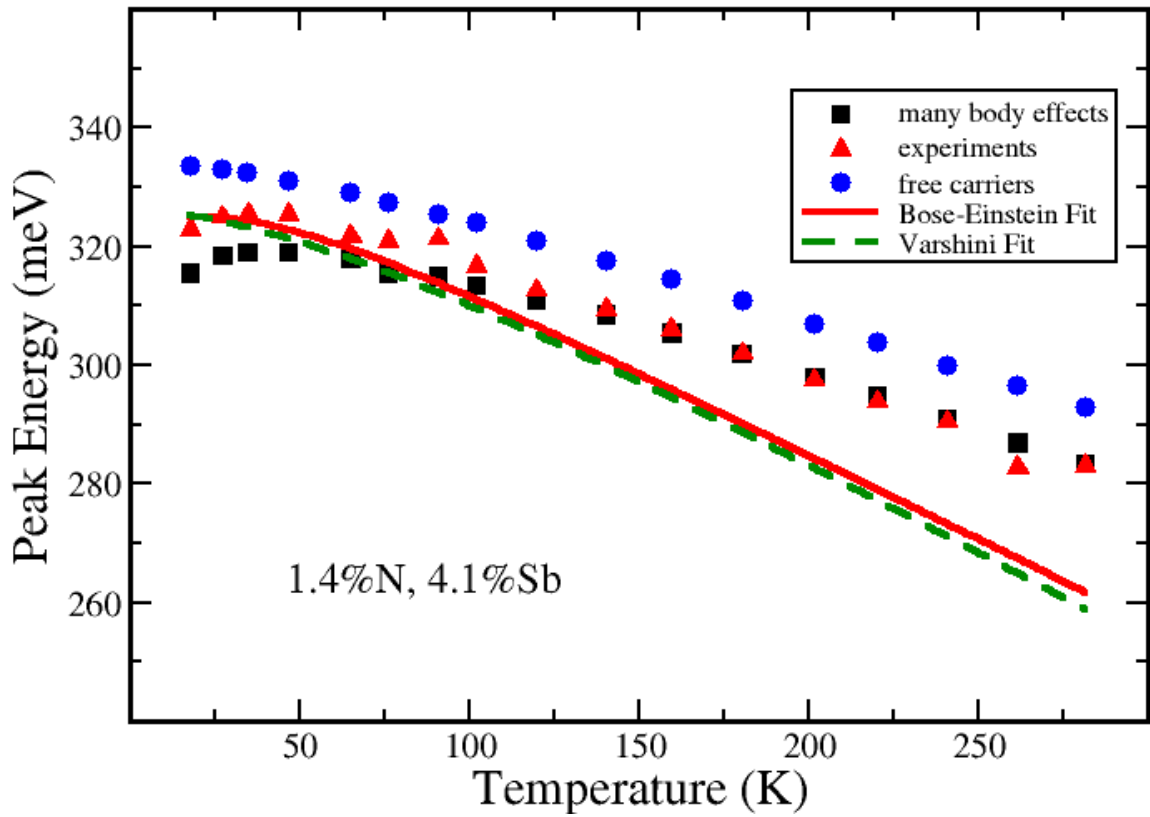


Fig. 5 Temperature dependence of the peak luminescence of for the mole fractions 1.4% N , 4.1% Sb . The black squares are is calculated with our many body approach, blue circles are for free carriers and the red triangles are experimental data extracted from Ref. [5] for comparison. The red solid and green dashed curves are the Bose-Einstein and Varshini fits for the bandgaps with parameters given in Ref. [5]. The Varshini and Bose-Einstein parameters of the Fit Equation (7) for the quaternary are given in Ref. [5]:

$$E_{g_0} = 326 \text{ meV}, \alpha = 0.328 \text{ meV} / \text{K}^2, \beta = 106 \text{ K}, E_{g_{0BE}} = 325 \text{ meV}, a_B = 20 \text{ meV}, \theta_B = 138 \text{ K}$$

The calculated emission peaks (black circles) with our approach containing many body effect are in good agreement with the experimentally measured luminescence energy emission peaks (red triangles) [5]. The blue circles in Fig. 5 depict the free carrier luminescence at different temperatures which dominate the entire spectrum when the bound state factor $g \rightarrow 0$. For both the calculated and experimentally observed luminescence, a blue shift is initially observed at low temperatures $\sim 10-80$ K before a gradual redshift at higher temperatures. The low temperature blue shift is due to exciton ionization, showing clearly the importance of our approach. The solid (red) and dashed (green) curves were calculated respectively with Bose-Einstein and Varshni fits to the band gap for a comparison. The strong deviations at large temperature further highlight the relevance of our approach and our choice of bandgap calculation, which reflects directly on the free carrier curve and the further improvement in agreement with experiments due to the combined effect of plasma screening, bandgap renormalization and excitonic enhancement in our many body calculations. The combination of the method presented here with other more fundamental bandstructure calculations beyond $\mathbf{k}\cdot\mathbf{p}$ [25] can lead to a very powerful predictive numerical tool to be used in the development of new materials and devices, notably dilute semiconductor lasers [26].

4 Conclusion

We have presented in this paper theoretical calculations of luminescence emission in dilute quaternary semiconductors of type $InAs_{1-x-y}N_xSb_y$. The bandstructure was obtained by applying the conduction and valence band anticrossing model on the nitrogen modified conduction band and the Sb modified valence band. Good agreement is obtained by comparing simulations and experimental findings of the evolution of spectra with different alloy compositions. The role of

the combined effect of plasma screening, bandgap renormalization and excitonic enhancement were demonstrated by comparing and contrasting calculations including these many body effects and the experiments against the free carrier case. The reason for including only these effects among further possibilities of other many body corrections is that they are sufficient to explain the experimental data, without introducing extra momentum dependence that would prevent the development of simple analytical solutions, which are a target for an approach that can be easily used by a large number of colleague are relatively easily programmed. Thus, this work has potential to support the numerical characterization of new semiconductor layers developed for devices targeting the mid infrared range.

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Biographies and photographs for the other authors are not available.

Caption List

Fig. 1 Band schematics of $InAs_{1-x-y}N_xSb_y$ dilute semiconductors.

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