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<th><strong>Title</strong></th>
<th>Modeling the optical constants of AlxGa1-xAs alloys</th>
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<tr>
<td><strong>Author(s)</strong></td>
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Abstract— Extension of Adachi's model with a Gaussian-like broadening function instead of a Lorentzian one is used to model the optical dielectric function of the alloy Al$_x$Ga$_{1-x}$As. Gaussian-like broadening is accomplished by replacing the damping constant in the Lorentzian line shape with a frequency dependent expression. In such a manner, the comparative simplicity of analytic formulae of the model is preserved, while the accuracy becomes comparable to more intricate models, and/or models with a significantly greater number of parameters. The employed model describes accurately the optical dielectric function in the spectral range from 1.5 to 6.0 eV in the entire alloy composition range. Relative rms error obtained for the refractive index is below 2.2% for all compositions.

I. INTRODUCTION

The alloy system Al$_x$Ga$_{1-x}$As/GaAs is of great technological importance in fabrication of various optoelectronic devices. Optical properties of solids are often described in terms of the complex optical dielectric function $\varepsilon(\omega) = \varepsilon_1(\omega) + \varepsilon_2(\omega)$. Problem with experimental data is that they are not expressed as continuous analytic functions of the electronic energy gaps and the alloy composition $x$. Therefore, it is required to model the experimental data with an analytical model [1-3]. The employed model must be simple and concise, and at the same time give reasonably good approximation of the optical spectra of investigated materials.

Adachi's model [4], [5] is relatively simple model which describes optical dielectric function with terms attributed to four energy gaps ($E_0, E_0+\Delta_0, E_1, E_1+\Delta_1$) and damped harmonic oscillators describing contributions from higher lying transitions ($E'_0, E'_2(X), E'_2(\Sigma)$). However, Adachi's model is not very accurate, and several modifications have been proposed recently [1], [6-10]. Jenkins [6] obtained better agreement with experimental data by introducing exponential decay of matrix elements which are taken to be constant over the Brillouin zone in Adachi's model. However, this model gives good agreement with experimental data only in the narrow range, and for AlAs calculated values differ from experimental ones for a constant amount below 3 eV. Zheng et al. [10] have recently proposed modification of Adachi's model which includes excitonic terms at $E_1$ and $E_1+\Delta_1$, gives new expression for the contribution of the indirect gap and introduces the Lorentzian lifetime broadening in a different manner. However, since model parameters (except the fundamental bandgap) are assumed to be linear functions of composition, this model does not yield the good agreement with experimental data for higher aluminum content ($x \geq 0.7$).

Kim et al. [11-13] have proposed an accurate but rather complicated model, which can include either Lorentzian or Gaussian broadening. Different types of broadening are accomplished by varying certain parameter in the expression for frequency dependent damping constant. The experimental data for Al$_x$Ga$_{1-x}$As in energy range from 1.5 eV to 6.0 eV and compositions from $x = 0.0$ to $x = 0.8$ in steps of 0.1 and $x = 1$ were fitted with 119 parameters. Obtained relative rms error for the refractive index using this model is below 2.5% for all compositions [12].

Model of Kim et al. is much more complex than the Adachi's model (and contains a large number of adjustable parameters), still the improvement in accuracy is obtained mainly through replacing the Lorentzian broadening function with Gaussian one. With simple Lorentzian broadening function this model doesn't provide the good agreement with experimental data in the vicinity of the direct edge and below. The fact that Lorentzian broadening doesn't accurately describe the optical spectrum has already been recognized and discussed [1], [11], [14], [15]. Rakić and Majewski [1] have shown that Adachi's model with Gaussian-like broadening function describes accurately dispersion and absorption in GaAs and AlAs even in the vicinity of the $E_0$ where the original model of Adachi is highly inaccurate.

In this paper we show that a comparatively simple model of Rakić and Majewski [1] can be successfully applied to model the optical spectrum of ternary alloys, in particular Al$_x$Ga$_{1-x}$As, with accuracy similar to that of significantly more intricate model of Kim et al. Also, we compare two ways of determining the parameters of the model for ternary compounds. First approach is to determine the model parameters for particular compositions, and then to find the optimal function describing the dependence of the model parameters on the alloy composition $x$. The second approach is to simultaneously fit the data sets for all available compositions in order to minimize the discrepancies between calculated and experimental data over the entire energy and composition range. Our results clearly show that simultaneous fitting was needed to provide accurate values of optical functions. Finally, we discuss what was the
advantage of using the global optimizing routine (namely our acceptance-probability-controlled simulated annealing algorithm [2, [3]]) compared to classical fitting algorithms, and how it affected the reliability of the final model parameters.

II. Model of the dielectric function

We shall briefly describe the applied model for the dielectric function. The dielectric function in Adachi's model is represented by the sum of terms describing transitions at critical points (CPs) in joint density of states. In the modification proposed by Rakić and Majewski [1] damping constants \( \Gamma_i \) are replaced with frequency dependent expression \( \Gamma_i(\omega) \).

A. \( E_0 \) and \( E_0 + \Delta_0 \) transitions

Under the parabolic band assumption, contributions of three-dimensional \( M_0 \) CPs \( E_0 \) and \( E_0 + \Delta_0 \) are given by

\[
e^I(\omega) = AE_0^{-3/2} \left[ f(\chi_0) + \frac{1}{2} \left( \frac{E_0}{E_0 + \Delta_0} \right)^{3/2} f(\chi_0) \right],
\]

where

\[
f(y) = y^{-2}[2 - (1 + y)^{1/2} - (1 - y)^{1/2}],
\]

\[
\chi_0 = \frac{\hbar \omega + i\Gamma_0}{E_0},
\]

\[
\chi_0 = \frac{\hbar \omega + i\Gamma_0}{E_0 + \Delta_0},
\]

where \( A \) and \( \Gamma_0 \) are strength and damping constant of the \( E_0 \) and \( E_0 + \Delta_0 \) transitions, respectively.

B. \( E_1 \) and \( E_1 + \Delta_1 \) transitions

For contributions of the two-dimensional \( M_0 \) CPs \( E_1 \) and \( E_1 + \Delta_1 \), by taking the matrix element to be constant with respect to energy, Adachi obtained the following expression

\[
e^{II}(\omega) = -B_1 \chi_1^{-2} \ln(1 - \chi_1^2) - B_{1s} \chi_1^{-2} \ln(1 - \chi_{1s}^2)
\]

where

\[
\chi_1 = \frac{\hbar \omega + i\Gamma_1}{E_1},
\]

\[
\chi_{1s} = \frac{\hbar \omega + i\Gamma_1}{E_1 + \Delta_1},
\]

\( B_1(B_{1s}) \) and \( \Gamma_1 \) are strength and damping constant of the \( E_1 \) and \( E_1 + \Delta_1 \) transitions, respectively. Contribution of the Wannier type 2D excitons (discrete series of exciton lines at the \( E_1 \) and \( E_1 + \Delta_1 \) CPs) is given by

\[
e^{III}(\omega) = \sum_{n=1}^{+\infty} \left( \frac{1}{(2n-1)^2} \frac{B_{1x}}{(E_1 - [G_1/(2n-1)^2] - \hbar \omega - i\Gamma_1} \right.
\]

\[
+ \left( \frac{2n-1)^2}{(E_1 + \Delta_1) - [G_1/(2n-1)^2] - \hbar \omega - i\Gamma_1} \right),
\]

where \( B_{1x} \) and \( B_{1s} \) are the strengths and \( G_1 \) and \( G_{1s} \) are Rydberg energies of \( E_1 \) and \( E_1 + \Delta_1 \) exciton, respectively. Here it was assumed that \( G_1 = G_{1s} = 0 \) [1]. Summation of the excitonic terms is performed until the contribution of the next term is less than \( 10^{-4} \).

C. \( E_0', E_2(X) \) and \( E_2(\Sigma) \) transitions

The origin of transitions \( E_0', E_2(X) \) and \( E_2(\Sigma) \) is not completely clear, since they do not correspond to a single, well defined CP. However, these features can be adequately modeled with damped harmonic oscillators, characterized with energy \( E_j \), oscillator strength \( f_j = \sqrt{C_j E_j^2} \) and damping constant \( \Gamma_j \), \( j = 2, 3, 4 \)

\[
e^{IV}(\omega) = \sum_{j=1}^{3} \frac{f_j}{E_j^2 - (\hbar \omega)^2 - i\hbar \omega \Gamma_j},
\]

D. The frequency dependent damping

Damping constants in equations (1)-(9) are replaced with

\[
\Gamma_i(\omega) = \Gamma_i \exp \left[ -\alpha_i \left( \frac{\hbar \omega - E_i}{\Gamma_i} \right)^2 \right].
\]

In this way, shape of the line varies with ratio of parameters \( \alpha_j \) and \( \Gamma_j \). Lineshapes range from purely Lorentzian (for \( \alpha = 0 \)) to nearly Gaussian (\( \alpha = 0.3 \)), while for large \( \alpha_j/\Gamma_j \) ratios wings of the peak in imaginary part of the dielectric function \( \varepsilon_2(\omega) \) are even narrower, thus enabling elimination of extended absorption tails in \( \varepsilon_2 \) which are characteristic for Lorentzian line shape. Since no broadening mechanism is set a priori (both \( \alpha_j \) and \( \Gamma_j \) are adjustable model parameters) model becomes very flexible.

E. Complete model for the dielectric function

Dielectric function is obtained by summing up the contributions of all the above described critical points, with \( \Gamma \) replaced by \( \Gamma(\omega) \)

\[
e(\omega) = \varepsilon_\infty + e^I(\omega) + e^{II}(\omega) + e^{III}(\omega) + e^{IV}(\omega),
\]

where \( \varepsilon_\infty \) is the high-frequency dielectric constant containing the contributions of higher lying transitions.

III. Results and discussion

Position of the \( E_0 \) and \( E_0 + \Delta_0 \), \( E_1 \) and \( E_1 + \Delta_1 \) and their variation with composition \( z \) is accurately determined in the study of Kim et al. [12] The energies of these critical points are given with

\[
e_i(x) = E_i(0) + (E_i(1) - E_i(0))x + (c_0 + c_1 x)z(1 - x),
\]

where values of \( E_i(0), E_i(1), c_0 \) and \( c_1 \) are listed in Table I. Therefore, energies of these CPs do not represent

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( E_i(0) )</th>
<th>( E_i(1) - E_i(0) )</th>
<th>( c_0 )</th>
<th>( c_1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( E_0 )</td>
<td>1.410</td>
<td>1.585</td>
<td>0.2242</td>
<td>-1.4235</td>
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<tr>
<td>( E_0 + \Delta_0 )</td>
<td>1.746</td>
<td>1.455</td>
<td>0.1931</td>
<td>-1.2160</td>
</tr>
<tr>
<td>( E_1 )</td>
<td>2.926</td>
<td>0.962</td>
<td>-0.2124</td>
<td>-0.7850</td>
</tr>
<tr>
<td>( E_1 + \Delta_1 )</td>
<td>3.170</td>
<td>0.917</td>
<td>-0.0734</td>
<td>-0.5093</td>
</tr>
</tbody>
</table>

TABLE I

Parameters describing composition dependence of four lowest critical points \( E_0, E_0 + \Delta_0, E_1 \) and \( E_1 + \Delta_1 \) according to Kim et al. [Phys. Rev. B 47, 1876 (1993)].
polynomial after estimating parameters for each composition separately. This was already pointed out by Terry [16], but most other computational efforts are demanding on the optimization algorithm and also more computationally intensive. The number of data points was larger approximately by an order of magnitude, and at the same time the number of fitting parameters was increased four times. Nevertheless, this effort can be justified from the number of points. Firstly, the model parameter estimation does not give single, unique solution of the problem. Similar quality of approximation of experimental data can be obtained with different sets of parameters.

Another problem is illustrated in Fig. 1. This figure depicts two different calculated curves. Solid line is a result of best simultaneous fit across all material compositions. Broken line is showing an interesting drawback of individual fitting. It is obtained by using cubic polinomial (cubic polinomial fit to parameters determined in individual fits). The descrepancies between the broken line and experimental data obviously reflect the difference between the cubic polynomial obtained in individual and simultaneous fits. It clearly shows the deterioration of the fit quality if optimal cubic polinomial is found after estimating parameters for each composition separately. Since main aim of modeling the optical properties of a ternary alloy is to enable calculation of the optical constants for compositions for which there are no available experimental data, it is clear that simulatenous approach to model parameter estimation for ternary alloy should be favoured. Therefore, in this work - results obtained by simultaneous fit to all compositions, dashed line - results corresponding to model parameters calculated by optimal cubic polinomial obtained by fitting each composition separately.
tain in this work, due to greater flexibility of the model achieved by frequency dependent damping mechanism and inclusion of the excitonic effects, can be clearly observed. In results presented here we didn’t include contributions of the indirect transitions since they exist only for $x > 0.45$ and their strength, since they represent second-order perturbation, should be significantly less. Fig. 3 shows the real and imaginary parts of the index of refraction vs. energy for compositions 0.3, 0.7 and 1.0. Obtained relative rms errors for the refractive index for our model with a total of 88 parameters are the largest for $x = 0.099$ and $x = 0.804$ (equal to 2.2%) and the lowest for $x = 0.315$ and $x = 0.419$ (equal to 1.4%). This is comparable to results of Kim et al. [12], who obtained rms errors below 2.5% with their model with 119 parameters, and results of Terry et al. who obtained rms errors below 3% with oscillator-based model with 144 parameters.

IV. CONCLUSION

Optical properties of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ are modeled in the 1.5-6 eV range for all compositions $0 \leq x \leq 1$. Extension of Adachi’s model employing adjustable broadening function instead of the conventional Lorentzian one is used. This article discusses the significance of employing the experimental data for all compositions simultaneously for model parameter estimation. It is shown that this approach yields to more accurate and reliable results than those obtained by employing cubic polynomial for approximation of the composition dependence of parameters obtained by fitting each composition separately. Excellent agreement with experimental data, illustrated by relative rms error for refractive index below 2.2%, is obtained in the entire investigated spectral range and for all compositions. Total of 88 adjustable model parameters is employed, which is significantly lower compared to other studies giving similar quality of approximation of optical constants of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ in the 1.5-6 eV range.

REFERENCES