

Theory of alloy broadening of deep levels in semiconductor alloys: Nitrogen in $\text{Al}_x\text{Ga}_{1-x}\text{As}$

William C. Ford and Charles W. Myles

Department of Physics and Engineering Physics, Texas Tech University, Lubbock, Texas 79409

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The inhomogeneous alloy broadening of deep levels produced by substitutional impurities in crystalline semiconductor alloys is investigated with use of a technique based upon the embedded-cluster method and the theory of deep levels. Results are presented for the composition dependences of the center, width, and component lines of the alloy-broadened spectrum of nitrogen on the arsenic site in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. For the present application, only the effects of the alloy disorder in the nearest-neighbor environment of the impurity are accounted for by the theory. In principle, however, the method outlined here is generalizable to account for such effects in the impurity's second- and more-distant-neighbor environments.

I. INTRODUCTION

The technological importance of the III-V compound semiconductor alloys is well established and has resulted in numerous experimental and theoretical studies of their electronic properties. The basic physics of these materials is also very interesting, because the random distribution of atoms in these systems can produce physical phenomena which do not occur in elemental or compound semiconductors. In particular, the alloy disorder in the neighborhood of defects in these materials produces fluctuations of the local potential energy from one defect site to another. Different defects of the same type within such an alloy can thus be surrounded by different chemical environments. This results in an effect where a given type of defect can produce numerous deep levels in the band gap, one for each possible distinct configuration of alloy constituents which can surround it. The deep-level spectrum for that type of defect can thus be inhomogeneously broadened by this mechanism. In recent years, several workers have observed and studied these disorder-broadening ("alloy-broadening") effects in optical absorption and luminescence from excitons bound to impurities in the III-V compound semiconductor alloys.¹⁻⁷ These studies have shown that the potential-energy fluctuations which broaden the impurity spectrum can also produce local environments which immobilize and hold excitons for relatively long times as they hop from a given defect site to one with lower energy, thus inhibiting their transport to traps and increasing the luminescence efficiency of the host material.

Although phenomenological theories of alloy-broadened luminescence spectra exist,^{8,9} and although there have been coherent-potential approximation (CPA) treatments⁵ of the alloy broadening of deep-level spectra, as well as one-^{10,11} and three-dimensional¹² model calculations of this effect, to our knowledge there is no published theory of the effect of alloy disorder on deep levels which both utilizes realistic band structures and accounts for local chemical environment effects in a realistic manner. In this paper, we outline a calculational technique which is

designed to overcome these deficiencies. It is based upon the embedded-cluster method¹⁰⁻¹⁴ and the Hjalmarson *et al.*¹⁵ theory of deep levels and it utilizes well-tested band structures¹⁶ as input. The embedded-cluster method is well established and has successfully treated a number of problems in alloy physics.¹⁰⁻¹⁴ Furthermore, the Hjalmarson *et al.*¹⁵ theory has been applied to numerous deep-level defect problems in semiconductors and has been successful in its predictions of the chemical trends for the deep levels for a large number of defect-host systems.¹⁷ Other theories of deep levels in semiconductors exist¹⁸⁻²⁰ and the approach we outline here could, in principle, be modified for use with any one of them. In the present work, however, our primary interest is the development of a technique which is capable of obtaining estimates of the alloy-disorder-induced, inhomogeneous broadening of deep levels in semiconductor alloys and of obtaining predictions of the trends in this broadening as a function of both the alloy composition and the defect. The Hjalmarson *et al.* theory¹⁵ is ideally suited for use with our technique because of its previous numerous successes in the predictions of such trends.¹⁷ Furthermore, despite its seeming simplicity, this version of deep-level theory has often given predictions which are in semi-quantitative agreement both with experiment¹⁷ and with more sophisticated calculations.²¹

As a particular application of our method, we consider here the effect of the nearest-neighbor disorder on the deep defect energy levels produced by a nitrogen impurity on the As site in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. However, the method we outline below is general and can, in principle, be applied to investigate alloy-disorder effects on the deep-level spectra of any defect in any semiconductor alloy. Applications to other impurities in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ and to defects in other semiconductor alloys will be discussed in a later paper.²²

II. METHOD

In our theory, the average properties of the alloy host are described using the nearest-neighbor sp^3s^* semiempir-

ical tight-binding band structures of Vogl *et al.*¹⁶ in the virtual-crystal approximation (VCA). It should be pointed out that, although the results we present below were obtained using these band structures and this alloy effective medium, our formalism is independent of the choice of both the band-structure model and the effective medium. We utilize the sp^3s^* band structures¹⁶ because they have proven accurate and useful in numerous applications to both defect¹⁷ and bulk²³ problems. The VCA effective medium has been chosen as the starting point for our description of the alloy host, rather than using the more complicated, self-consistent CPA medium²⁴ (as was done in most of the previous applications of the embedded-cluster method¹⁰⁻¹⁴) because the alloy of interest here, $\text{Al}_x\text{Ga}_{1-x}\text{As}$, is known to have a relatively small disorder parameter. It thus falls into the category of alloys known as weak scattering systems,²⁴ for which the VCA is expected to be an adequate effective-medium representation.²⁴ Coherent-potential approximation calculations carried out for this alloy by Chen and Sher²⁵ and Shen and Myles²⁶ (the latter calculations used as input the same band-structure model as is used here) confirm this expectation, yielding CPA self-energies of the order of a few hundredths of an eV and producing densities of states which are virtually indistinguishable from those calculated using the VCA. It should be pointed out, however, that in applying our method to other semiconductor alloys, the VCA effective medium should be used with caution and that the more correct CPA medium should certainly be used for any alloy with a large disorder parameter.

In our approach, the local properties of the alloy are described by employing the embedded-cluster method developed by Gonis and Garland¹⁴ and Myles and Dow.¹³ Specifically, a five-atom cluster consisting of a central anion (As) tetrahedrally surrounded by its four nearest-neighbor cations (either Ga or Al) in a particular configuration is embedded in the VCA medium. The impurity (N) atom is then placed on the central anion site of the cluster, and its persistent deep level (if it is in the band gap) is found with use of the Hjalmarson *et al.*¹⁵ theory of deep levels. This procedure is repeated for each distinct cluster configuration and the contribution of each cluster's deep level to the overall spectrum is weighted by the probability of occurrence of that configuration.

Formally, this procedure is carried out as follows. First, the virtual-crystal Green's function²⁴

$$g = (E - H_v)^{-1} \quad (1)$$

is defined, where H_v is the sp^3s^* Hamiltonian of Vogl *et al.*¹⁶ for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ calculated in the VCA, and E is an energy. For a particular cluster configuration, a cluster Green's function G for the five-atom cluster described above is calculated, following the embedded-cluster method,^{13,14} by solving the Dyson equation

$$G = g + gVG = (1 - gV)^{-1}g, \quad (2)$$

where V is a cluster potential defined as

$$V = H_{\text{cl}} - H_v \quad (3)$$

for sites inside the cluster and which vanishes outside the

cluster. Here H_{cl} is the sp^3s^* Hamiltonian¹⁶ for the particular cluster configuration under consideration. Finally, the deep levels for that configuration for an impurity on the As site of the cluster are obtained using the theory of deep levels.¹⁵ This involves solving the Fredholm determinantal equation

$$\det[1 - G(E)U] = 0, \quad (4)$$

where U is the defect potential describing the impurity.

For an sp^3 -bonded substitutional impurity such as that considered here, U is a 4×4 matrix (one s and three p orbitals) so that Eq. (4) is a 4×4 determinant. In obtaining the results described below, we have followed most of the major assumptions of the Hjalmarson *et al.* deep-level theory.¹⁵ In particular, in solving Eq. (4), we assume that U is a diagonal matrix with the diagonal s -orbital matrix element equal to U_s and the diagonal p -orbital matrix elements each equal to U_p . This assumption neglects lattice relaxation in the vicinity of the impurity. We also utilize Hjalmarson's empirical rule¹⁵ that U_p is equal to one-half U_s . These assumptions are consistent with previous applications^{15,17,21} of the Hjalmarson *et al.* theory,¹⁵ where they were justified for developing global theories for predicting chemical trends. Inclusion of lattice-relaxation effects, which would require U to be nondiagonal, and not implementing the empirical relation between U_p and U_s are both possible using the present approach. However, these modifications to the theory, while increasing its accuracy for specific defects, would require significantly more computational effort with the resultant sacrifice in the global aspects of the theory.

With these assumptions for U , and for fixed energy E , Eq. (4) then becomes a quartic algebraic equation for the s -orbital defect potential energy U_s . From the four solutions to this equation, as functions of E , the deep-level energy versus defect potential energy diagrams, which are one of the common results of the Hjalmarson *et al.* theory, are generated.¹⁵ One of these solutions gives the s -like deep level (which is usually labeled A_1 for isolated impurities in nonalloyed semiconductors) and three of them are the p -like deep levels. In what follows, for convenience, we consider the s -like solution only. The p -like solutions are qualitatively similar to those we present below.

The above procedure is repeated for each distinct cluster configuration, resulting in an impurity state density spectrum of the form

$$D(E) = \sum_i P_i \delta(E - E_i), \quad (5)$$

where i labels the configurations,²⁷ P_i is the probability of occurrence of the i th configuration,²⁸ E_i is the deep level in the band gap found for configuration i , and the sum on i goes only over configurations which produce a deep level in the band gap. Moments of this spectrum are easily computed.¹⁰ Here we consider only the first moment and the second moment about the mean or the width. It should be pointed out that while we assume here that the alloy is completely random so that P_i is a binomial distribution,²⁷ this assumption affects only the moments computed via Eq. (5) and not the component levels obtained

from Eq. (4). Nonrandomness and short-range order in the alloy host could be taken into account in our approach by appropriately modifying our choice of the probability distribution P_i , which would in turn alter our results for the first moment and the width. In this regard, it should be noted that recent experimental²⁹ and theoretical³⁰ studies have shown that in many of the ternary semiconductor alloys, the distribution of nearest-neighbor bond lengths is bimodal, which indicates a nonrandom occupation of the lattice sites and a difference in the lengths of the cation-anion bonds between the alloy constituents.

In order to specify a particular impurity to study, it is necessary to determine U_s , which fixes the deep-level energy for each cluster configuration from the E vs U_s diagram for that configuration. In the Hjalmarson *et al.* theory,¹⁵ U_s is determined for a particular impurity by using Hjalmarson's empirical rule¹⁵ that it is proportional to the difference in the s -orbital atomic energies of the impurity and the host atom it replaces. As is well known, this assumption enables the prediction of the trends in the level depths as a function of both impurity and host and it has successfully made such predictions for numerous defect-host systems.¹⁷ However, the absolute level depth one obtains from this assumption is not always in agreement with that found experimentally. Since, in the present paper we are interested in obtaining as accurate a prediction as possible of the trends in the effects of alloy disorder on the nitrogen defect energy level in $\text{Al}_x\text{Ga}_{1-x}\text{As}$, and since, as will be discussed in a later publication,²² the amount of disorder broadening predicted by our theory depends strongly on the depth of the deep level, for the results presented here we have taken an alternate approach which enables us to obtain an estimate of the broadening which is unobscured by inaccuracies in the level depth. Specifically, we have treated the potential U_s as an empirical parameter which we adjust so that the nitrogen deep level in AIAs predicted by the theory agrees exactly with that found experimentally. This value of U_s is then used in the alloy-broadening calculations outlined above. This is not a very radical departure from the assumptions of Hjalmarson *et al.*,¹⁵ where U_s is also treated as an empirical parameter. It merely alters the empirical rule by which this quantity is determined. Furthermore, lattice-relaxation effects, omitted from the Hjalmarson *et al.*¹⁵ theory, are at least partially included in this manner because the deep level at $x = 1$ is fit to the experimental value, which of course contains these effects. The experimental nitrogen-level depth in AIAs is $E = 2.218$ eV (Ref. 31) (0.0518 eV below the conduction-band edge), which gives a theoretical value of $U_s = -2.5566$ eV when U_s is adjusted so that the theoretical depth in AIAs agrees with the experimental one. This is the fixed value of U_s which we use to determine the nitrogen depth for all alloy compositions and cluster configurations.

III. RESULTS AND DISCUSSION

The results for the alloy-broadened deep-level spectrum obtained for the nitrogen impurity on the As site in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ by the procedure outlined above are summa-

rized in Figs. 1 and 2. In Fig. 1, we show the alloy composition dependence of the component deep levels for each of the five unique nearest-neighbor cluster configurations which can surround the impurity.²⁷ Also shown in that figure is the composition dependence of the average energy. In Fig. 2 we show the composition dependence of the width of the spectrum.

It is clear from Fig. 1 that all of the component energy levels are independent of the conduction-band edge as x is varied, as is characteristic of deep levels in general. Furthermore, these levels vary almost linearly with x and, unlike calculations of such levels made using perturbation theory,³² they are not equally spaced. This unequal level spacing is attributable to the differences in the off-diagonal matrix elements of the sp^3s^* Hamiltonian for AIAs and GaAs,¹⁶ which are included as off-diagonal matrix elements of the cluster potential V , Eq. (3). Such effects are not included in usual perturbation-theory calculations.³²

The reasons for the order in which the component levels become conduction-band resonant (as a function of decreasing x) can be understood qualitatively by using a "defect-molecule" argument, following Hjalmarson.¹⁵ In particular, consider the three Al and one Ga cluster as x decreases from unity and the host acquires more GaAs character. Detailed comparisons of the energy-level structures of a GaAs "host molecule" and an AlN "defect molecule" show that the deep levels produced by the defect molecule should be conduction-band resonant, in agreement with the actual cluster calculation.²² Similar considerations hold for the other clusters' deep levels. More details will be presented in a later paper.²²

The behavior of the first moment of the spectrum may be understood by considering the behavior of the component levels whose configuration average produce it. As the different configuration's deep levels exit the band gap (as x decreases from unity) and become conduction-band resonant, the first moment is suddenly displaced deeper into the band gap due to the fact that the levels which

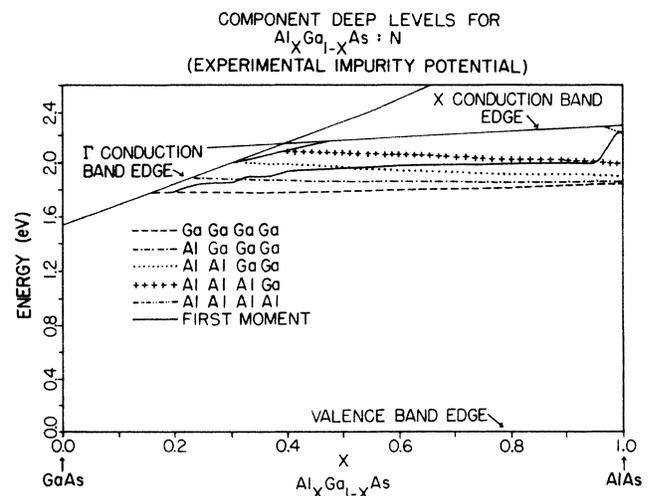


FIG. 1. Composition dependence of the component lines and the average energy for the disorder-broadened deep-level spectrum of N in $\text{Al}_x\text{Ga}_{1-x}\text{As}$.

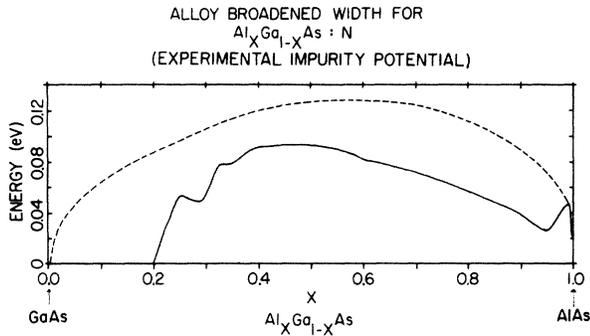


FIG. 2. Composition dependence of the width of the disorder-broadened deep-level spectrum of N in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ if only component deep levels in the band gap are included (solid curve) and if both band-resonant levels and levels in the band gap are included (dashed curve).

have become band resonant no longer contribute to this quantity. (Band-resonant levels, which cannot under ordinary circumstances be observed, do not contribute to the spectrum we have computed and are thus given a weight of zero.) Except at compositions where a component deep level becomes band resonant, the first moment closely follows the VCA deep-level energy within the error of the sp^3s^* tight-binding parameters.¹⁶ This implies that, at least for $\text{Al}_x\text{Ga}_{1-x}\text{As}$, the center of the deep-level spectrum is relatively insensitive to the theory one uses to characterize the alloy host material.

In Fig. 2, we illustrate the computed results for the linewidth as a function of x for the same case as for Fig. 1. In that figure, the solid curve indicates the linewidth computed as described above, with only the component deep levels which lie in the band gap contributing. As with the first moment, the shape of this curve can be understood by noting that a dip occurs at each composition where a component deep level (as a function of decreasing x) becomes conduction-band resonant. On the other hand, the dashed curve indicates the expected linewidth profile if both band resonant and, in the gap, cluster deep levels are included. Of perhaps more importance than the shape of the linewidth vs x curve is its magnitude, which is a measure of the expected magnitude of the alloy broadening. For nitrogen in $\text{Al}_x\text{Ga}_{1-x}\text{As}$, the present theory predicts that this width is of the order of 40–90 meV for most compositions. Thus, the broadening of deep levels due to nearest-neighbor alloy disorder could in some cases be a non-negligible and even an important effect. It is also important to note that the alloy-broadened linewidth is not symmetric about $x=0.5$, which is in marked contrast to perturbation theory³² and model calculation approaches.¹²

Finally, some comments concerning the experimental observability of the effects we predict here are in order. Since recent time-resolved luminescence experiments on excitons bound to nitrogen in $\text{Ga}_x\text{In}_{1-x}\text{P}$ (Ref. 8) have

resolved component deep-level lines, it is reasonable to speculate that similar experiments would have a reasonable chance of observing such effects for nitrogen in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. Furthermore, the linewidths we predict for the overall spectrum are certainly well within the experimentally observable range. However, in analyzing data, it should be borne in mind that effects such as lattice relaxation, which are not included in the present theory, can contribute to the observable linewidth and cause it to deviate from that which we predict. Also, in a time-resolved experiment, as in Ref. 8, dynamical effects such as differing capture cross sections for the component levels and temperature-dependent level occupations can both tend to narrow the luminescence spectrum in comparison with the random linewidth we compute here.⁹

In summary, we have presented a technique for calculating the inhomogeneous alloy broadening of deep levels produced by substitutional impurities in semiconductor alloys. As an example, we have applied it to predict the composition dependences of the center, width, and component lines of the alloy-broadened spectrum of nitrogen on the arsenic site in $\text{Al}_x\text{Ga}_{1-x}\text{As}$. Although only the effects of the alloy disorder in the nearest-neighbor environment of the impurity are accounted for in the present application, the theory is, in principle generalizable to account for such effects in the impurity's second- and more-distant-neighbor environments. The method is also, in principle, generalizable to account for lattice-relaxation and short-range order effects which can also contribute to observed widths.

The principle findings of the present paper are the following: (1) Both the first moment and the component levels are independent of the conduction-band edge, as is characteristic of deep levels, (2) the behavior of the first moment and the linewidth as a function of x can be understood by noting the compositions at which the component levels become conduction-band resonant (as a function of decreasing x), (3) the magnitude of the alloy broadening for N in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ is of the order of 40–90 meV, as measured by the rms width, and (4) the level splittings are of the order of 10 meV. The latter two facts indicate that the effects we predict here should be observable with modern optical techniques.

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