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# Dependence of the energy and form of the optical-absorption onset of interface diffusion in $Si_nGe_m$ superlattices

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The optical absorption coefficient $\alpha(E)$ of $Si_nGe_m$ superlattices depends strongly on the quality of the Si-Ge interfaces. Interdiffusion across the Si-Ge interfaces is reflected on the variation of $\alpha(E)$ with energy for energies near the energy gap $E_g$. Using the coherent potential approximation for simulating the interdiffusion across the Si-Ge interfaces and the Kubo-Greenwood formula for the optical absorption, we show that for $Si_5Ge_5$, $\alpha(E)\approx(E-E_g)^\kappa$ where the exponent $\kappa$ takes values in the range 0.5–2.0 depending on the degree of interdiffusion across the Si-Ge interfaces. The value $\kappa=0.5$ reflects abrupt Si-Ge interfaces while the value $\kappa=2$ corresponds to highly diffused Si-Ge interfaces. The case $\kappa=2$ fits very well the available experimental data. [S0163-1829(97)50608-4]

Recent experimental results reported on the optical properties of the $Si_nGe_m$ strain-symmetrized superlattices (SS-SL's) have indicated that their explanation cannot be justified by model approximations which assume an abrupt Si-Ge interface. In particular, Olajos, *et al.*[1] have measured the optical absorption coefficient $a(\hbar\omega)$ for the $Si_nGe_n$ SS-SL's for $n=5,6$ and found that at the onset (i.e., for energies very close to the energy gap, $E_g$) it exhibits a variation,

$$a(\hbar\omega)\propto(\hbar\omega-E_g)^\kappa \tag{1}$$

with $\kappa=2$, which cannot be explained by assuming an abrupt Si-Ge interface. Further evidence that supports the presence of nonabrupt Si-Ge interfaces has been deduced from other results as well. For example, the observation that the measured band gap of the $Si_3Ge_2$ SS-SL's was found identical to that of the corresponding $Si_{0.6}Ge_{0.4}$ alloy[2] suggests the existence of interface mixing. Moreover, the similarity in the electroreflectance spectra of $Si_6Ge_4$ and $Si_4Ge_6$ found by Asami *et al.*,[3] as well as the experimental evidence of thermal instability of SL's,[4] indicate considerable interface diffusion. Furthermore, the measured phonon spectra of the $Si_nGe_n$ SS-SL's (Ref. 5) could not justify abrupt Si-Ge interfaces. Finally, the mixed ordering of Si and Ge atoms at the Si-Ge interfaces of their corresponding SS-SL's has been confirmed in the works of Jesson, Pennycook, and Baribeau,[6] and Müller *et al.*[7]

The interdiffusion and mixing in the Si-Ge interface as well as their effect on the optoelectronic properties of the $Si_nGe_m$ SS-SL's has recently attracted the efforts of various theoretical studies.[8–13] The theoretical investigations have indicated that, on the one hand, interdiffusion across the Si-Ge interface results in intermixed interfaces (ordered or randomized) that are more stable than the abrupt (nonintermixed or noninterdiffused) Si-Ge interfaces.[13] On the other hand, theory has shown that the diffused interface weakens the optical absorption coefficient of the SS-SL's as compared to the corresponding one of the system which exhibits abrupt Si-Ge interfaces.[8–12] Weakening of the optical absorption coefficient was also found but to a lesser extent, in the case of SS-SL's with ordered intermixing at the Si-Ge interfaces.[8]

The weakening of $a(\hbar\omega)$ in the case of diffused interfaces may be well understood by considering the changes the interdiffusion imposes on the band structure of $Si_nGe_m$ SS-SL's. These SL's, being of type II, exhibit a band structure in which the valence band is mostly determined by the Ge layers and the conduction band of the Si layers. As a result of the interdiffusion, the valence band will change in the direction of approaching the Si valence band (therefore lowering its energy) while the conduction band will change in the direction of approaching the conduction band of Ge (therefore increasing its energy). We have demonstrated in a recent paper[11] that, indeed, interdiffusion widens the energy gap in the case of the $Si_5Ge_5$ SS-SL and as a result weakens the optical absorption coefficient. Such a result has been reported by other investigators as well.[8,10,12] However, the success of the theory to give quantitative results comparable to experiment depends on the degree of computational accuracy which in turn depends on the efficiency of the theory to describe sufficiently the interdiffusion (or the mixing) across the Si-Ge interface. In the work of Turton and Jaros,[8] this requirement is achieved by performing calculations using local empirical pseudopotentials and employing a supercell containing 8000 atoms. Using an empirical tight-binding model Hamiltonian with an $sp^3$ basis set but restricted to a supercell containing only 40 or 90 atoms, Theodorou and

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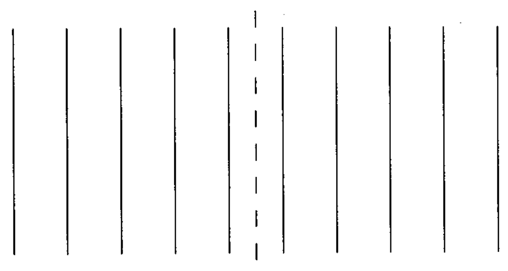
Tserbak[10] calculated the dependence of $a(\hbar\omega)$ on the photon energy $\hbar\omega$ for energies near the band gap $E_g$; they found that for a randomness 50% in the Si and Ge lattice planes nearest to the interface, and 25% randomness on the second planes nearest to the interface, the square root of the absorption coefficient is approximately proportional to the energy for the $Si_5Ge_5$, $Si_6Ge_4$, $Si_4Ge_6$ SS-SL's (for energies near $E_g$) and claimed agreement with the experimental findings.[1] In a completely different approach, we have shown[11] that the optical properties of SS-SL's with diffused Si-Ge interfaces can be more efficiently and very accurately studied using the Kubo-Greenwood[14] formula for the optical absorption by incorporating to this the effect of randomness within the coherent potential approximation (CPA).[15] This approach, originally suggested by Pickett, Papaconstantopoulos, and Economou,[16] allows one to include transitions in the optical-absorption process that allow a violation of crystal momentum conservation within an uncertainty in the wave vector **k** that is imposed by the degree of randomness. In the case of the $Si_nGe_m$ SS-SL's, the degree of randomness is specified by the degree of interdiffusion and mixing of the Si-Ge interfaces.

However, although the reported theoretical investigations agree quite well on the qualitative description of the effect of the interface disorder on the optical-absorption coefficient, they fail to give quantitative results in agreement with experiment. In particular, the theoretical optical-absorption coefficient is found two orders of magnitude smaller than the experimental one[12] and the theoretical variation with frequency of $a(\omega)$ near its onset has been found to vary according to Eq. (1) with the exponent $\kappa > 1$, contrary to the experimental value $\kappa = 1$ for the $Si_nGe_m$ SS-SL's with $n \approx m$. Partial agreement between theory and experiment seems to be recovered[1,10,12] in the case of $Si_nGe_n$ SS-SL's with $n = 5,6$. Thus, the complete verification of Eq. (1) remains an open problem which requires more elaborate investigations.

In the present paper, we present a systematic analysis based on an accurate calculational approach which allows one to study the energy and the form of the optical-absorption coefficient $a(\hbar\omega)$ near its onset, as well as its dependence on the quality of the interfaces. The proposed analysis will help to interpret the existing experimental data and make predictions for further work.

In our model, the randomness in the Si-Ge interfaces is introduced as follows:[11] Each plane parallel to the interface is considered as a random substitutional alloy of $Si_{x_i}Ge_{1-x_i}$ type characterized by its silicon concentration $x_i$, $i = 1, \ldots, N$, where $N$ is the number of the lattice planes of the SL within its unit cell. In Fig. 1 a schematic representation of the $Si_5Ge_5$ SL is shown with silicon concentrations (along the $N = 10$ lattice planes) suitable to describe an abrupt and a diffused Si-Ge interface. Following Turton and Jaros,[8] we define the degree of randomness $d$ in the Si-Ge interface as twice the number of misplaced atoms divided by the total number of atoms.

In the present work, we investigate the effect of the interdiffusion across the Si-Ge interfaces on the energy dependence of the optical-absorption coefficient of the $Si_5Ge_5$ SS-SL grown along the $\langle 001 \rangle$ direction on a $Si_{0.56}Ge_{0.44}$ amorphous substrate. For this SS-SL, the effect of zone folding is to create a direct energy gap at the $\Gamma$ point.[11] It will be shown that for energies very close to the energy gap $E_g$, the optical-absorption coefficient $a(\hbar\omega)$ exhibits a variation with the photon energy $\hbar\omega$ dictated by Eq. (1) with the exponent $\kappa$ depending strongly on the degree of randomness $d$. In particular, it will be shown that $\kappa \approx \frac{1}{2}$ in the case where the Si-Ge interface is abrupt $(d = 0)$; this is characteristic of absorption connected with momentum-conserving transitions between parabolic energy bands if the oscillator strength is assumed constant.[17] As the interdiffusion is allowed to take place in the Si-Ge interface, it will be shown that the exponent $\kappa = 2$ in the case of highly diffused Si-Ge interfaces $(d > 0.5)$. This case is characteristic of indirect transitions in materials with parabolic dispersion of energy bands and results, in the present case, from the randomness across the interface which relaxes the momentum conservation in the optical transition processes.



| $d$ | $x_1$ | $x_2$ | $x_3$ | $x_4$ | $x_5$ | $x_6$ | $x_7$ | $x_8$ | $x_9$ | $x_{10}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 0.00 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 0.16 | 0.8 | 1.0 | 1.0 | 1.0 | 0.8 | 0.2 | 0.0 | 0.0 | 0.0 | 0.2 |
| 0.32 | 0.7 | 0.9 | 1.0 | 0.9 | 0.7 | 0.3 | 0.1 | 0.0 | 0.1 | 0.3 |
| 0.48 | 0.6 | 0.8 | 1.0 | 0.8 | 0.6 | 0.4 | 0.2 | 0.0 | 0.2 | 0.4 |

FIG. 1. Side view of the unit cell of the $Si_5Ge_5$ SL. Solid lines represent the lattice planes parallel to the interface. Below each plane the disorder parameter $d$ and the corresponding Si concentration $x_i$ on each plane are indicated for the cases studied.

Our calculational approach is based on the following basic steps: (1) the tight-binding (TB) calculational method which is used to determine the band structure of the SL in an efficient and accurate way. This is achieved by employing the three-center integral approximation and the Hamiltonian TB matrix elements corrected in order to include strain effects.[9] (ii) The CPA within its TB scheme,[15] which is used for simulating the interdiffusion across the Si-Ge interfaces. This is achieved by describing each random alloy $Si_{x_i}Ge_{1-x_i}$, associated with the $i$th lattice plane parallel to the interface, with a coherent potential function $\Sigma_j(x_i;E)$, $j = s, p_x, p_y, p_z$, as obtained by applying the CPA.[11] (iii) The Kubo-Greenwood theory,[14] which is used to obtain the optical absorption coefficient $\alpha(\hbar\omega)$ of the SL. More details of our method are given in Refs. 11 and 18.

In our calculations the geometry of the SL remains *"diamondlike"* with the lattice constants parallel and perpendicular to the interface being different because of the strains induced by the substrate and the interfaces. Valence-band offsets are taken from Ref. 19 corrected for strain effects by interpolation.[9] In the case of the diffused interfaces, the diagonal matrix elements of our TB Hamiltonian are replaced by the coherent potentials $\Sigma_j(x_i;E)$. The so obtained TB Hamiltonian $H_{\Sigma(E)}$ is used to obtain the Green function from the relation $G(E)=[E-H_{\Sigma(E)}]^{-1}$, which in turn is used to study the optical properties of the SL by calculating the optical conductivity $\sigma(\hbar\omega)$ according to the formula of Kubo and Greenwood:[14]

$$\sigma(\hbar\omega)=\frac{2e^2}{\pi\Omega m^2\omega}\lim_{\delta\to 0}\sum_{\mathbf{k}}\int_{E_F-\hbar\omega}^{E_F}dE \times\mathrm{Tr}\{\tilde{p}_x \mathrm{Im}[G(\mathbf{k},E+i\delta)]\tilde{p}_x \times\mathrm{Im}[G(\mathbf{k},E+\hbar\omega+i\delta)]\}, \quad (2)$$

where $\tilde{p}_x=d\tilde{H}(\mathbf{k})/dk_x$ is the momentum matrix ($\tilde{H}$ denotes the virtual crystal TB Hamiltonian) and $\delta$ a real positive constant. Having $\sigma(\hbar\omega)$ we obtain the optical absorption coefficient $a(\hbar\omega)$ from the relation $a(\hbar\omega)=4\pi\sigma(\hbar\omega)/nc$ where $c$ is the speed of light and $n$ the index of refraction (in our calculations we have taken $n=1$).

The determination of the exponent $\kappa$ in Eq. (1) from calculations that employ Eq. (2) is numerically a very difficult step. The major difficulties are associated with the **k** integration and the process of taking the limit $\delta\to 0$ in Eq. (2). The latter step is necessary because even very small values of $\delta$ introduce artificial electron density of states within the band gap which in turn introduce significant errors in the evaluation of the integral over the energy. The smaller the value of $\delta$ the greater the number of **k** points that is needed. For example, for $d=0.00$ and for $\delta=0.0025$ eV, as set of 17 000 **k** points in the irreducible part of the Brillouin zone is necessary for the **k** integration in Eq. (2).

In order to investigate the effect of the interdiffusion across the Si-Ge interfaces on the optical absorption and its variation with energy for energies very close to the energy gap, we have calculated the optical conductivity and the optical-absorption coefficient using Eq. (2) for various degrees of randomness $d$. Subsequently, we expressed our results for $a(\hbar\omega)$ according to the variation indicated by Eq. (1) and we found the variation $\kappa=\kappa(d)$. In Fig. 2 we present our results for the variation of the energy gap (direct gap at $\Gamma$— see inset of Fig. 2) with the degree of randomness $d$. The variation of $\kappa=\kappa(d)$ has been found to be as follows: $\kappa$=0.50, 0.66, 1.54, 2.10 for $d$=0.00, 0.16, 0.32, 0.48, respectively, the values of $d$ corresponding to the Si depth profiles shown in Fig. 1. Finally, in Fig. 3 we present the onset optical-absorption coefficient versus frequency for $d=0.0$ and 0.32. As it is apparent from the band structure of $\mathrm{Si}_5\mathrm{Ge}_5$, shown in the inset of Fig. 2, the main contributions to $\alpha(\omega)$ near its onset arise from direct transitions at the $\Gamma$ point (shown by the arrow $VC$).

Prior to commenting on our results, it is worth noticing that the present results, as those reported in Refs. 8–12, suffer from the limitations of the independent electron approach. As a result of these limitations, the $E_1$ peak of the calculated imaginary part of the dielectric function, $\epsilon_2(\omega)$, is underestimated (especially for Si) while its $E_2$ peak is usually overestimated. For a better description of $\epsilon_2(\omega)$ [and therefore for $\alpha(\omega)$ since $\alpha(\omega)=\epsilon_2(\omega)\omega/n(\omega)c$], one has to include excitonic effects which are not taken into account in the present work (see, for example, Ref. 20 and references therein).

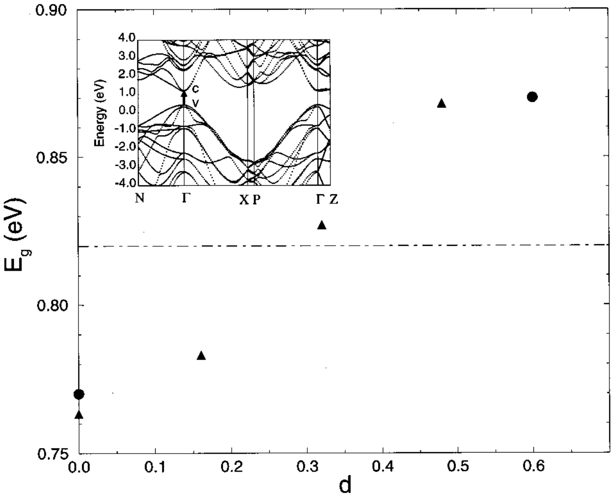


FIG. 2. Results of the present work for the variation of the energy gap $E_g$ at $\Gamma$ (see inset) with the disorder parameter $d$ (triangles). The results of Theodorou and Tserbak (Ref. 10) are shown with circles. The horizontal line corresponds to the experimental value interpolated appropriately in order to fit the theoretical period length of the SL. The main contribution to the optical-absorption coefficient $\alpha(\omega)$ near its onset arises from transitions at the $\Gamma$ point (arrow $VC$ in the inset, where the band structure for $d=0$ is shown).

Our calculations were restricted to the values $d=0.00$, 0.16, 0.32, and 0.48. These values of $d$ correspond to configurations of Si concentration along the lattice planes parallel to the interface (and within the unit cell) shown in Fig. 1. Our results for $d=0.00$ (abrupt interface) and for $d\approx 0.50$ (strong interdiffusion) exhibit the expected behavior for transitions between parabolic energy bands that conserve or not the momentum, respectively. However, our results for the ideal SL, $(d=0)$, do not agree with those reported recently in Refs. 10 and 12 which report $\kappa=1$. Concerning this discrepancy, we observe that the results of Refs. 10 and 12 are not consistent with the energy bands of $\mathrm{Si}_5\mathrm{Ge}_5$ SS-SL's, which are found parabolic around the $\Gamma$ point and the fact

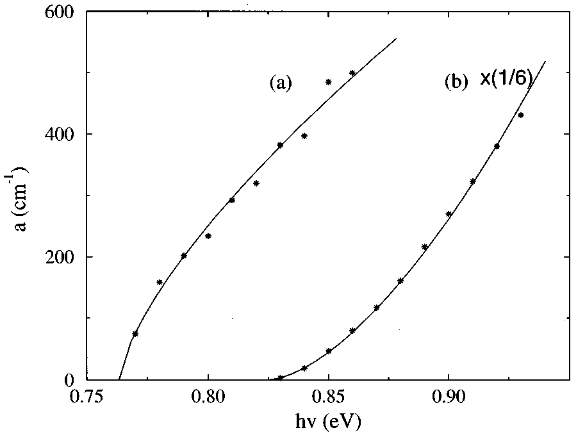


FIG. 3. The absorption coefficient versus photon energy for $\delta=0.0025$ and disorder parameter (a) $d=0.00$ and (b) $d=0.32$. The dots correspond to the calculated values, while the solid lines represent the fitted curve $\alpha(\hbar\omega)=\mathrm{const}\times(\hbar\omega-E_g)^{\kappa}$ with (a) $\kappa=0.5$ and (b) $\kappa=1.6$.

that the only contributions to the optical-absorption coefficient at the onset region arise from transitions around the $\Gamma$ point. This can be directly verified by calculating the spectral representation of the electron density of states. Therefore, for $d=0$, it seems more reasonable to conclude that the exponent $\kappa$ is equal to 0.5 as it is found in our calculations and that the experimental findings of Pearsall *et al.*[12] correspond to SL's with diffused Si-Ge interfaces. In addition to the mentioned discrepancy, our results for $\alpha(\omega)$, near its onset for $d=0$, are larger (by a factor of 3) as compared with those of Refs. 10 and 12. Unfortunately, the majority of the existing experimental data for $\alpha(\omega)$ are given in arbitrary units. Thus, a direct comparison with experiments is not possible. However, Pearsall *et al.*[12] report that the experimental values of $\alpha(\omega)$ (for SL's with $|m-n|=1$) may be much larger than their theoretical results. If this is true for the present case as well (for which $m=n$), our results would be closer to the experiment than those of Refs. 10 and 12. On the other hand, the results of Theodorou and Tserbak[10] corresponding to $d=0.60$, are in very good agreement with ours. Although there is no unique correspondence between the degree of randomness $d$ and the model configurations for Si concentration within the unit cell, we do not expect major qualitative differences in our results that depend on the configurations $x_i$, $i=1,\ldots,N$. This is because the randomness across the Si-Ge interfaces relaxes the momentum conservation in the optical transitions and, combined with the square root densities of states exhibited approximately by our system, leads to $\kappa\approx 2$ in Eq. (1) as the randomness is increased.

Our method allows us also to calculate matrix elements for the optical transitions by dividing the result of Eq. (2) by the convoluted density of states calculated at the same frequency $\omega$. From these calculations[18] we find that the transition matrix elements (averaged over both polarizations) indicate a strong dependence on the disorder parameter droping about two orders of magnitude as $d$ increases from zero to 0.48 in good agreement with the results of Turton and Jaros.[8]

In conclusion, we have shown that randomness due to interdiffusion across the Si-Ge interfaces of the $Si_5Ge_5$ SS-SL's can be reflected in the variation of the optical-absorption coefficient at the frequency onset region. In the case of SL's which exhibit a direct energy gap and parabolic energy bands, this variation can be described by Eq. (1) where the exponent $\kappa$ depends on the degree of randomness $d$ increasing from $\kappa=0.5$ (when $d=0$) to $\kappa\approx 2.0$ for large $d$. Although a universal relation $\kappa=\kappa(d)$ may not exist, nevertheless, the present investigation gives a reasonable representation for this variation. It is understood that, in the case of SL's, which do not exhibit direct energy gap, the onset frequency variation of $\alpha(\hbar\omega)$ does not follow necessarily the behavior obtained in this work. Thus the results of the recent work by Engvall *et al.*,[21] indicating a $\kappa>1$, do not imply a diffuse interface. Within the present analysis and in the absence of experimental information about the Si (or Ge) depth profile in the examined SL's, it can be concluded that although the reported experimental data[1,12] do not allow yet a direct comparison with the theory, they, however, give strong evidence of interface mixing.

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