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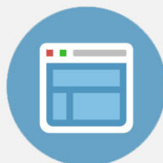
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Optical properties of SiGe alloys

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The optical properties of $Si_{1-x}Ge_x$ have been investigated theoretically using a full-potential linear muffin-tin-orbital method. We present the density-of-states as well as the real and imaginary parts of the dielectric function. The calculated dielectric function was found to be in good agreement with the spectroscopic ellipsometry measurements by J. Bahng *et al.*, J. Phys.: Condens. Matter **13**, 777 (2001), and we obtained a static dielectric constant of $\epsilon_0 = 12.19 + 2.45x$ in the Si rich regime ($x \leqslant 0.5$). © *2003 American Institute of Physics.* [DOI: 10.1063/1.1555702]

I. INTRODUCTION

$Si_{1-x}Ge_x$ is a good candidate as a substitute material for Si in low-power and high-speed semiconductor device technologies.[1,2] Optoelectronic devices, such as heterojunction bipolar transistors (HBT),[3,4] are already in industrial production. $Si_{1-x}Ge_x$ is also promising as alloying material for quantum well devices,[5] infrared detectors,[6] and modulation-doped field-effect transistors (MODFET).[7,8] Although much effort has been paid to the growth of $Si_{1-x}Ge_x$ and $Si_{1-x}Ge_x$/Si as well to electrical characterization, there is still a lack of information about the optical properties of $Si_{1-x}Ge_x$.

In this article, the electronic structure of $Si_{1-x}Ge_x$ has been calculated, using a first-principle full-potential linear muffin-tin-orbital method (FPLMTO).[9] We present the total and the partial density-of-states as functions of composition x. The imaginary part of the dielectric function was calculated from the joint density-of-states and the optical matrix elements, whereupon the real part of the dielectric function was obtained by using the Kramer–Kronig dispersion relation. We show that the calculated dielectric functions are in very good agreement with spectroscopic ellipsometry measurements, recently presented by J. Bahng *et al.*[10]

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II. COMPUTATIONAL METHOD

In order to study the electronic structure of $Si_{1-x}Ge_x$ we have used the FPLMTO method,[9] which is based on the local-density approximation (LDA) with the Hedin–Lundqvist[11] parametrization for the exchange and correlation potential. The unit cell was constructed with the eight atoms conventional unit cell, obtained from a fcc lattice with a two-point basis. The composition was varied by changing the atom type of the eight atoms, yielding $x=0$, 1/8, 2/8, ..., 1. The lattice constant for the different compositions was obtained from an optimization of the total energy. The basis functions, the electron densities, and the LDA potentials were calculated with the geometrical optimized volume. These quantities were expanded in combinations of spherical harmonic functions (with a cutoff $\ell_{max}=6$) inside nonoverlapping spheres surrounding the atomic sites (muffin-tin spheres) and in a Fourier series in the interstitial region. The muffin-tin spheres occupied ~60% of the unit cell. The radial basis functions within the muffin-tin spheres are linear combinations of radial wave functions and their energy derivatives, computed at energies appropriate to their site and principal as well as orbital atomic quantum numbers, whereas outside the muffin-tin spheres the basis functions are combinations of Neumann or Hankel functions.[12,13] In the calculations reported here, we made use of valence band $3s$, $3p$, and $3d$ basis functions for Si, pseudo-core $3d$ and valence band $4s$, $4p$, and $4d$ basis functions for Ge and, valence band $2s$, $2p$, and $3d$ basis functions for C with

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corresponding two sets of energy parameters. The resulting basis formed a single, fully hybridizing basis set. This approach has previously proven to give a well converged basis.[9] To sample the irreducible wedge of the Brillouin zone we used the special **k**-point method.[14] We used 216 **k** points in the irreducible Brillouin zone. We checked the convergence of the total energy with respect to the number of **k** points. In order to speed up the convergence we have associated each calculated eigenvalue with a Gaussian broadening of width 136 meV.

III. CALCULATION OF THE DIELECTRIC FUNCTION

The ($\mathbf{q}=\mathbf{0}$) dielectric function was calculated in the momentum representation, which requires matrix elements of the momentum, **p**, between occupied and unoccupied eigenstates. To be specific the imaginary part of the dielectric function, $\epsilon_2(\omega)\equiv \mathrm{Im}\,\epsilon(\mathbf{q}=\mathbf{0},\omega)$, was calculated from[15]

$$\epsilon_2^{ij}(\omega)=\frac{4\pi^2e^2}{\Omega m^2\omega^2}\sum_{\mathbf{k}nn'\sigma}\langle\mathbf{k}n\sigma|p_i|\mathbf{k}n'\sigma\rangle\langle\mathbf{k}n'\sigma|p_j|\mathbf{k}n\sigma\rangle \times f_{\mathbf{k}n}(1-f_{\mathbf{k}n'})\delta(e_{\mathbf{k}n'}-e_{\mathbf{k}n}-\hbar\omega). \quad (1)$$

In Eq. (1), e is the electron charge, m its mass, Ω is the crystal volume and $f_{\mathbf{k}n}$ is the Fermi distribution. Moreover, $|\mathbf{k}n\sigma\rangle$ is the crystal wave function corresponding to the nth eigenvalue with crystal momentum **k** and spin σ.

With our spherical wave basis functions, the matrix elements of the momentum operator are conveniently calculated in spherical coordinates and for this reason the momentum is written $\mathbf{p}=\Sigma_\mu \mathbf{e}_\mu^* p_\mu$,[16] where μ is -1, 0, or 1, and $p_{-1}=(p_x-ip_y)/\sqrt{2}$, $p_0=p_z$, and $p_1=-(p_x+ip_y)/\sqrt{2}$.[17]

The evaluation of the matrix elements in Eq. (2) is done over the muffin-tin region and the interstitial space separately. The integration over the muffin-tin spheres is done in a way similar to what Oppeneer[18] and Gasche[15] calculated using the atomic sphere approximation (ASA). A full description of the calculation of the matrix elements was presented elsewhere.[19] In our theoretical method the wave function [Eq. (1)] inside the muffin-tin spheres is atomic-like in the sense that it is expressed as a radial component times spherical harmonic functions (also involving the so called structure constants), i.e.,

$$\Psi_{mt}^{\bar{k}}(\bar{r})=\sum_t c_t\chi_t^{\bar{k}}, \quad (2)$$

where the sphere R', the basis function is

$$\chi_t^{\bar{k}}=\frac{\Phi_t(\bar{r}-\bar{R})}{\Phi(S_{mt}^R)}\delta_{\bar{R},R'}-\sum_{t'}\frac{\Phi_{t'}(\bar{r}-\bar{R}')}{\Phi(S_{mt}^{R'})}S_{t,t'}^{\bar{k}}, \quad (3)$$

and

$$\Phi_t(\bar{r})=i^lY_l^m(\hat{r})[\varphi(r)+\omega(D)\dot{\varphi}(r)]. \quad (4)$$

In Eqs. (2), (3), and (4) S_{mt}^R is the muffin-tin radius for atom R, $S_{t,t'}^{\bar{k}}$ is the structure constant, D is the logarithmic derivative, $\varphi(r)$ is the numerical solution to the spherical component of the muffin-tin potential and $\dot{\varphi}(r)$ is the energy derivative of $\varphi(r)$.[12,13] Therefore, this part of the problem is quite analogous to the ASA calculations[15,18] and we calculate the matrix elements in Eq. (2) as

$$\langle\mathbf{k}n\sigma|p_\mu|\mathbf{k}n'\sigma\rangle=\sum_{t,t'}c_tc_{t'}^*\langle\chi_{t'}|p_\mu|\chi_t\rangle. \quad (5)$$

Since, χ_t involves a radial function multiplied with a spherical harmonic function, i.e., $f(r)Y_l^m$ (we will label this product $|l,m\rangle$), we can calculate the matrix elements in Eq. (5) using the relations

$$\langle l+1,0|p_0|l,0\rangle=\frac{l+1}{\sqrt{(2l+1)(2l+3)}}\times\left\langle f^*(r)\left(\frac{\delta}{\delta r}-\frac{l}{r}\right)f(r)\right\rangle, \quad (6)$$

and

$$\langle l-1,0|p_0|l,0\rangle=\frac{l}{\sqrt{(2l-1)(2l+1)}}\times\left\langle f^*(r)\left(\frac{\delta}{\delta r}+\frac{l+1}{r}\right)f(r)\right\rangle. \quad (7)$$

A general matrix element, $\langle l',m'|p_i|l,m\rangle$ is then calculated using the Wigner–Eckart theorem.[16] Matrix elements of the momentum over the interstitial are obtained from the relation

$$\int_{\Omega_{\mathrm{int}}}d^3r(\psi_i^*\nabla\psi_j)=\frac{1}{\kappa_j^2}\int_{\Omega_{\mathrm{int}}}d^3r(\psi_i^*\nabla^2\nabla\psi_j). \quad (8)$$

By use of Green's second theorem the expression above can be expressed as

$$\int_{\Omega_{\mathrm{int}}}d^3r*(\psi_i^*\nabla^2\nabla\psi_j)=-\int_{S_{MT}}dS\left(\psi_i^*\nabla\frac{d}{dr}\psi_j\right). \quad (9)$$

In the expression above, the surface integral is taken over the muffin-tin spheres and since the interstitial wave function by construction is the same as the muffin-tin wave function we can use for ψ the numerical wave function defined inside (and on the boundary of) the muffin-tin spheres. In this way the evaluation of the integral above is done in a quite similar way as done for the muffin-tin contribution [Eqs. (6)–(8)] to the gradient matrix elements.

The summation over the Brillouin zone in Eq. (1) is calculated using linear interpolation on a mesh of uniformly distributed points, i.e., the tetrahedron method. Matrix elements, eigenvalues, and eigenvectors are calculated in the irreducible part of the Brillouin zone. The correct symmetry for the dielectric constant was obtained by averaging the calculated dielectric function. Finally, the real part of the dielectric function, $\epsilon_1(\omega)$, is obtained from $\epsilon_2(\omega)$ using the Kramers–Kronig transformation:

$$\epsilon_1(\omega)\equiv\mathrm{Re}[\epsilon(\mathbf{q}=0,\omega)]=1+\frac{1}{\pi}\int_0^\infty d\omega'\;\epsilon_2(\omega')\times\left(\frac{1}{\omega'-\omega}+\frac{1}{\omega'+\omega}\right). \quad (10)$$

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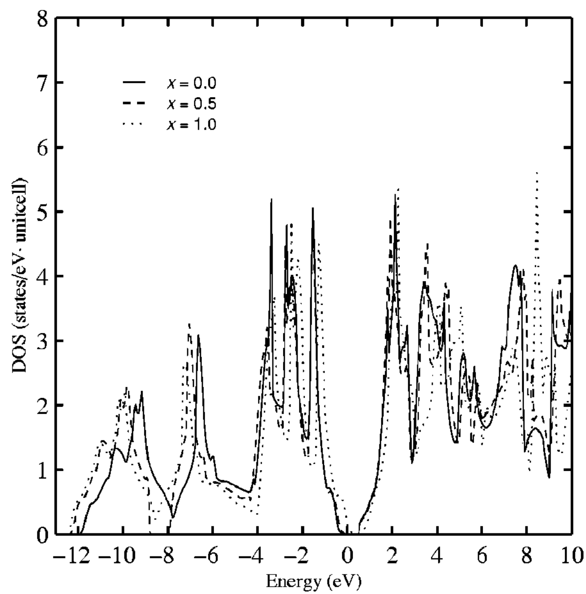
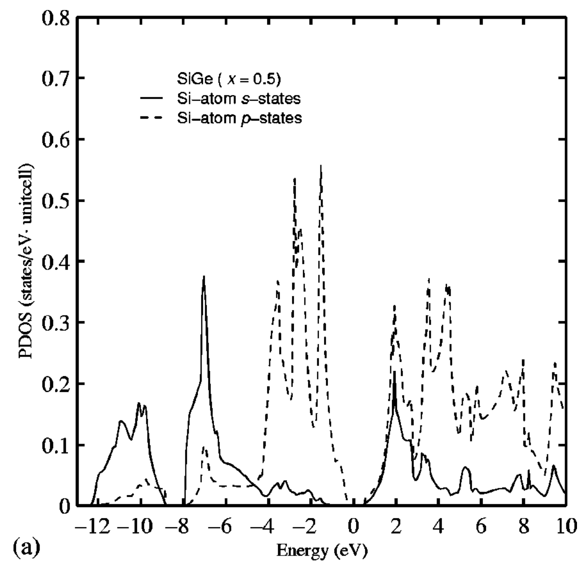


FIG. 1. Total density-of-states of $Si_{1-x}Ge_x$ for $x=0.0$ (solid line), $x=0.5$ (dashed line), and $x=1.0$ (dotted line).



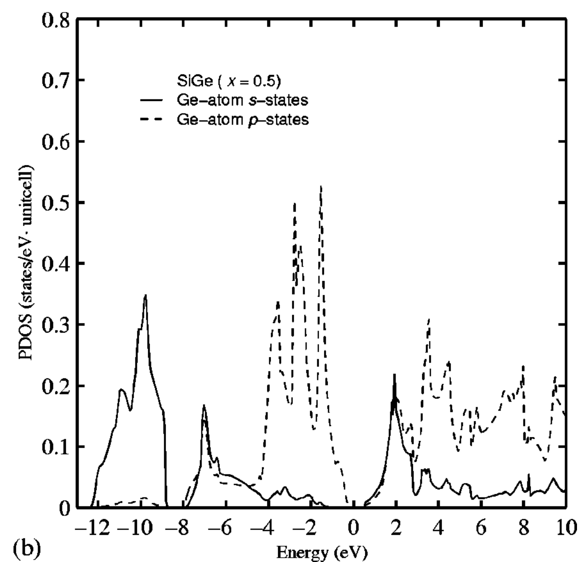


FIG. 2. Partial density-of-states of SiGe $(x=0.5)$ for a Si atom (a) and a Ge atom (b).

IV. RESULTS

The geometric optimization of the crystal structure gives a lattice constant:

$$a=5.393+0.237x \quad \text{Å}, \tag{11}$$

which obeys Végard's law of a linear behavior of the lattice constant of alloys.[20] Furthermore, the calculated result is close to the experimental data[21] $a=5.434+0.223x$ Å, although slightly underestimated which is normal within the LDA.

The calculated density-of-states of $Si_{1-x}Ge_x$ is shown in Fig. 1. The similarity of the density-of-states for different compositions is obvious. The main difference, however, is the larger valence-band width for Ge rich alloying. For instance, the total widths of the valence band of Si, SiGe (i.e., $x=0.5$), and Ge are 11.7, 12.3, and 12.8 eV, respectively.[22] The experimental values for Si and Ge are 12.5 and 12.7 eV, respectively. The LDA bandwidths for semiconductors are known to be underestimated.

The partial densities-of-states of SiGe $(x=0.5)$ are rather similar for the Si and the Ge atoms (see Fig. 2). The valence band consists of two subbands, where the lower lying subband (about -12.3 to -8.8 eV) is mainly s character, and the upper subband (-7.9–0 eV) has an s character in its lower energy regime and almost only a p character in its upper energy regime. The main difference between the density-of-states of Si and of Ge atoms, is that Si has more $2s$ character in the upper subband, whereas Ge has a very strong $3s$ character in the lower subband. Si also has a somewhat stronger p character in the low energy regime of the sp-hybridizated conduction band.

Figure 3(a) shows the spectroscopic ellipsometry measurement of the imaginary part $\epsilon_2(\omega)$ of the dielectric function performed by Bahng *et al.*[10] The peak at 4.2 eV was interpreted as interband transitions at the X point and along the Σ line. The corresponding calculated dielectric function is shown in Fig. 3(b), with a Lorentz broadening of 70 meV. Even if it is not straightforward to compare the density-of-states with the absorption spectrum, mainly due to the optical matrix elements, the strong peak at 3.5–3.75 eV can most likely be associated with transitions from the p-like states at about -2.0 eV in the valence band to the sp-hybridizated conduction band at about 1.5 eV. These energies originate from the uppermost valence band and the lowest conduction band in regions around the X and K points, and these transition energies are rather insensitive to the composition x. By including a band-gap correction of about 0.5 eV we identify the 3.5–3.75 eV peak with the measured peak of about 4.2 eV, which is in correspondence with Lautenschlager *et al.*[23]

The strong experimental low-energy peak at 2.2–3.5 eV, denoted E_1 and $E_1+\Delta_1$ in Ref. 10, depends strongly on the alloy composition. This peak was interpreted as interband transitions along the Λ line (Γ–L). For Si $(x=0)$ that is consistent with the transition energy across the band gap of

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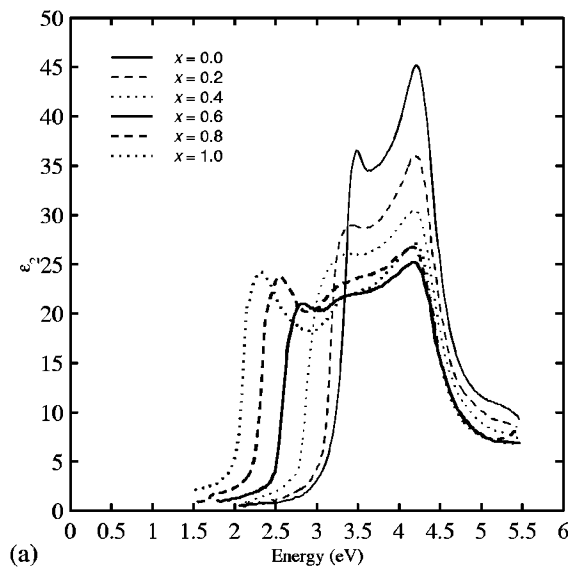

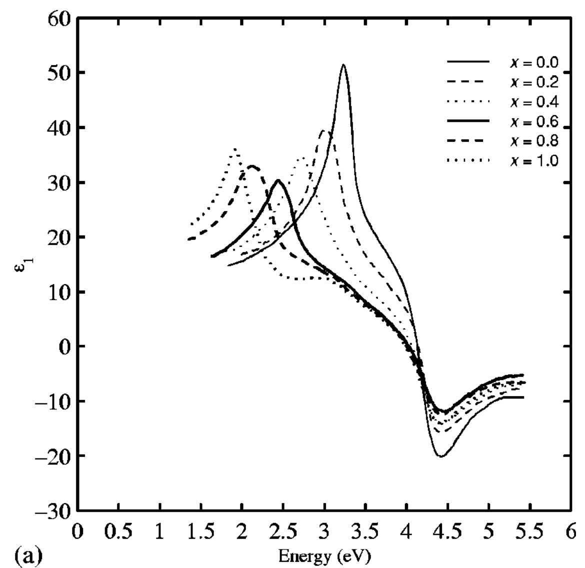

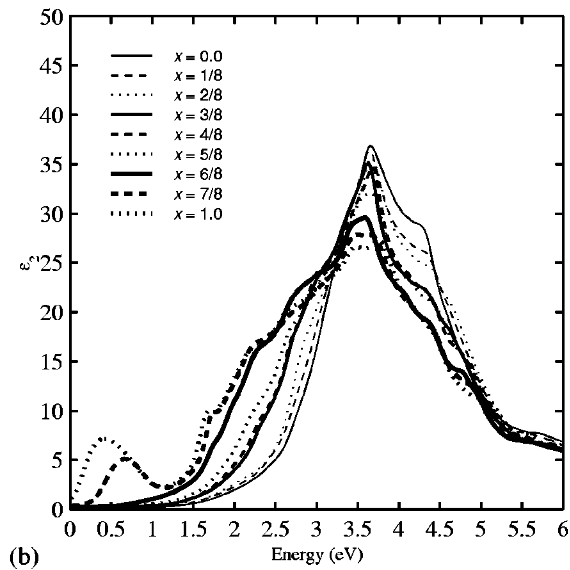

FIG. 3. Measured (a) and calculated (b) imaginary part $\epsilon_2(\omega)$ of the dielectric function.

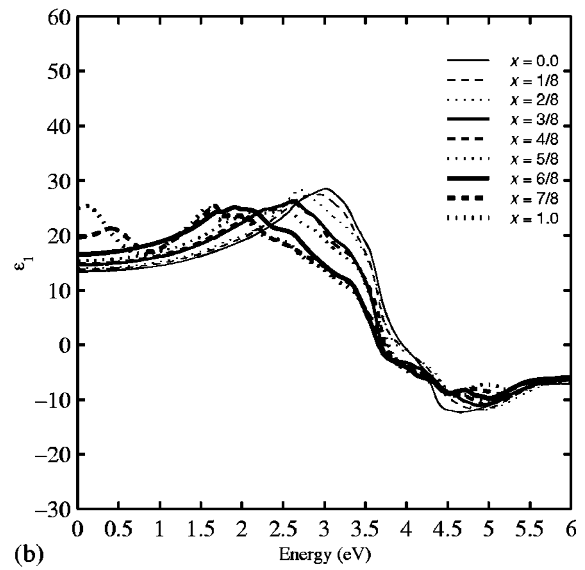

FIG. 4. Measured (a) and calculated (b) real part $\epsilon_1(\omega)$ of the dielectric function.

about 3.5 eV at the Γ point.[22] Our calculations also show that the energy band gap along the Λ line is fairly constant, which should result in strong absorption for this energy. Nevertheless, we do not obtain any sharp absorption peak, even if one can see a small tendency to a broad peak for Ge rich alloys associated with transitions along the Λ line. However, in Ref. 10 an excitonic line shape was assumed for fitting this sharp low-energy absorption peak. Therefore, it is not surprising that we cannot reproduce this sharp peak since the present LDA method is a ground state calculation and cannot describe excitonic states.

Figure 4 shows the real part of the dielectric function $\epsilon_1(\omega)$, where the calculated dispersions were obtained from the Kramer–Kronig dispersion relation. Overall, we find a very good qualitative agreement between the calculated and the measured results. The relatively stable negative peak at about 4.5 eV as well as the composition dependent positive peak at about 2–3 eV can be seen in both Figs. 4(a) and 4(b).

The polarity of the Si–Ge bond can with reasonable accuracy be neglected,[24] which means that the zero-frequency transverse optical and longitudinal optical modes are degenerate. This implies $\epsilon_1(0)\approx\epsilon_1(\omega>0)$ for $\omega\ll E_g/\hbar$. In order to calculate the static dielectric constant [i.e., $\epsilon_0\equiv\epsilon_1(0)$] accurately, we have included the band-gap correction (0.5 eV), yielding a constant shift in the absorption energy, whereas the real part of the dielectric function at low frequencies was calculated. This procedure has been shown to be the most proper way to include the band-gap correction in LDA calculations.[25]

From the FPLMTO calculation we obtained the static dielectric constant in the Si rich regime as $\epsilon_0=12.19+2.45x$, $x\leqslant 0.5$. For Si ($x=0$) this is close to the experimental value of $\epsilon_0=11.7–12.1$.[22] We cannot calculate the static dielectric constant for Ge rich alloys, since the cou-

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pling between the conduction sp-like band and the valence-band maximum with p-like states is overestimated within LDA, resulting in a too low band-gap energy for $x>0.85$. The effect on the dielectric function due to the strong coupling can be seen in Fig. 4(b) for $x=7/8$ and 1 for energies below 1 eV.

V. CONCLUSIONS

The optical properties of $Si_{1-x}Ge_x$ have been calculated by using the FPLMTO method. The calculated dielectric functions are in good agreement with measurements performed by J. Bahng *et al.*[10] We identify the 4.2 eV absorption peak as direct interband transitions in the Brillouin zone around the X and K points. We cannot observe the sharp measured low-energy absorption peak, which most likely is due to exciton effects. The static dielectric constant was found to be $\epsilon_0=12.19+2.45x$ for $x\leqslant 0.5$, which for Si is in very good agreement with the experimental value.[22]

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